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International Conference  
on  
Aerosols and Background Pollution

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## Abstracts

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University College, Galway,  
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13 - 15 June, 1989

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**AEROSOLS AND BACKGROUND POLLUTION CONFERENCE**

University College Galway 1989

**ABSTRACTS**

**Sponsors and Supporters**

The European Association for the Science of Air Pollution  
The European Research Office, London  
The Gesellschaft fur Aerosolforschung  
The Aerosol Society  
Western Regional Tourism Organization Ltd.  
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University College Galway

CONFERENCE PROGRAMME

AEROSOLS AND BACKGROUND POLLUTION

Tuesday 13 June

0830 Registration

0945 Official Conference Opening

**SESSION 1:**

**AEROSOL CLIMATOLOGY**

1000 Aerosol Climatology : invited review  
O. Preining

1045 Refreshment break

1115 Development of a global-scale background aerosol model  
D.A. Bowdle

1130 Arctic Haze: Long range pollution of the polar atmosphere  
R.C. Schnell and J.D. Kahl

1145 Southern Ocean condensation nuclei and climate variations  
J.L. Gras

1200 Cloud condensation and Aitken nuclei measurements in background air  
E.K. Bigg, A.D. Kaye and W.J. Megaw

1215 Aerosol climatology measurements with Nolan-Pollak counters  
T.C. O'Connor and F.M. McGovern

1230 Lunch

**SESSION 2:** MARINE AEROSOLS

1345 Marine Aerosols : invited review  
J.W. Fitzgerald

1430 Marine Aerosols generated from bursting air bubbles  
J. Wu

1445 Summertime aerosol measurements in the Ross sea region of Antarctica.  
M.J. Harvey, I.S. Lechner, G.W. Fischer and P. Isaac

1500 Physical characteristics of the ambient aerosol at Mace Head  
S.G. Jennings, C.D. O'Dowd, T.C. O'Connor and F.M. McGovern

1515 North Atlantic Aerosol Background concentrations measured at a Hebridean coastal site  
M.H. Smith, P.M. Park and I.E. Consterdine

1530 Refreshment break

1600           **SESSION 2 continued**

1600           Physical and chemical parameters of marine background and  
                  Sahara aerosols over the North Atlantic ocean  
                  V. Dreiling, R. Maser and L. Schutz

1615           Measurements of aerosol concentration and distribution at  
                  Helgoland Island  
                  P. Brand, J. Gebhart, M. Below, B. Georgi and J. Heyder

1630           Volatility of background aerosol  
                  S.G. Jennings and C.D.O'Dowd

1645           Interhemispheric variability in aerosol physicochemistry over  
                  the remote Pacific ocean  
                  A.D. Clarke

1700           Comparison of aerosol characteristics in the arid southwestern  
                  US and the west coast of western Europe  
                  E.H. Holt, R.G. Pinnick and S.G. Jennings

1715           Distribution of methanesulfonate, NSS-sulfate and  
                  Dimethylsulfide over the Atlantic and the North Sea  
                  S. Burgermeister and H. -W. Georgii

1800           Reception - Aula Maxima

2100           Irish style evening - Salthill Hotel

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Wednesday 14 June

SESSION 3: PROPERTIES OF AEROSOLS

0900 The chemistry of aerosols in the atmosphere: invited review  
R.M. Harrison

0945 Elemental composition of aerosol particles under background conditions in Hungary  
I. Borbély-Kiss, E. Kolai, E. Mészáros, Á. Molnár and Gy. Szabó

1000 Particulate and gaseous air pollutant levels at the Portuguese west coast  
C. A. Pio, R.M. Leal, I.M. Santos and T.D. Anacleto

1015 Acid aerosol measurements at a suburban Connecticut site  
G. Keeler, J. Spengler, and R. Castillo

1030 Sensitivity limit, resolution, counting efficiency and coincidence limit of optical aerosol particle counters  
H.T. Sommer

1045 Refreshment break

1115 Session 3: PROPERTIES OF AEROSOLS (CARBONACEOUS) continued

1115 Characterization of the carbonaceous aerosol in the western Mediterranean atmosphere  
H. Cachiér, M.P. Bremond and P. Buat-Ménard

1130 Internal/external mixing of soot to aerosols in source and receptor regions in Austria  
R. Hitzenberger

1145 A methodology for estimating regional emissions of black carbon aerosol in the Arctic  
J.D. Kahl, A.D.A. Hanson and R.C. Schnell

1200 Airborne particulate elements carbon: its sources, transport and soiling potential  
R.S. Hamilton and T.A. Mansfield

1215 Particle samples from forest fires  
A.D. Kaye and W.J. Megaw

1230 Atmospheric aerosol sources and their climatic significance  
G.A. d'Almeida, P. Koepke and M. Hess

1245 Lunch

1345           **SESSION 3: PROPERTIES OF AEROSOLS (OPTICAL) continued**

1345           Atmospheric optical depths : background and perturbations in the troposphere and stratosphere  
**R.F. Pueschel, P.B. Russell and M.P. McCormick**

1400           Spectral extinction coefficients of background aerosols in Europe, North America and South America; A comparison  
**H. Horvath**

1415           Evidence of a tropospheric aerosol backscatter background mode at CO<sub>2</sub> wavelengths  
**J. Rothermel, D.A. Bowdle, R.T. Menzies, M.J. Post and J.M. Vaughan**

1430           The Mauna Loa aerosol backscatter intercomparison experiment (MABIE)  
**D.A. Bowdle, E.M. Patterson, M.J. Post and A.D. Clarke**

1445           Aerosol optical properties in remote marine regions  
**A.D. Clarke**

1500           Refreshment break

1530           **POSTER PAPERS PRESENTATION**

**SESSION 1 : AEROSOL CLIMATOLOGY**

Continuous background aerosol monitoring with the epiphanimeter  
**U. Baltensperger, H.W. Gaggeler and D.T. Jost**

A thermo-optic technique for in-situ analysis of size-resolved aerosol physicochemistry  
**A.D. Clarke**

Background aerosol measurements in Germany  
**B. Georgi**

Comparison of aerosol size distribution measurements in an urban and remote site in Switzerland  
**B. Giorgi, H. -N. Müller and M. Meurer**

**SESSION 3: PROPERTIES OF AEROSOLS (CHEMICAL):**

Reaction of monoterpenes with ozone,  $\text{SO}_2$  and  $\text{NO}_2$  gas phase oxidation of  $\text{SO}_2$  and formation of sulphuric acid  
**D. Kotzias, K. Fytianos and F. Geiss**

Atmospheric conditions of photochemical smog formation in cities  
**O.V. Lomaya and D.F. Kharchilava**

Terpenes, ozone and nitrogen oxide in a Finnish background station  
**T. Raunemaa and S. Konttinen**

Case studies of the chemical composition of atmospheric aerosol at the Szrenica summit, Karkonosze range, Poland  
**J.W. Zwozdziak**

**PROPERTIES OF AEROSOLS (PHYSICAL):**

Simulation of aerosol particles in a large smog chamber  
**H. Bunz, M. Koyra and W. Schock**

A novel instrument for the classification of air borne particles on the basis of size, shape and count frequency  
**N.A. Eyles, P.H. Kaye and I.K. Ludlow**

Sampling artifacts from curved airborne intake systems  
**B.J. Huebert, W. Warren and G. Lee**

A study of particulate resuspension from an ash-dump  
**J.S. Pastuszka, J. Dybiczanski and M. Borowska**

Application of an isothermal haze chamber to studies of mixed soluble/insoluble anthropogenic particles  
**C.F. Rogers**

Microphysical characteristics evolution of the natural aerosol just before fog formation  
**R. Serpolay**

**PROPERTIES OF AEROSOL (OPTICAL):**

Periodicity of extinction curve of dielectric spherical particles  
**Peter Chýlek, J. Zahá, and S.G. Jennings**

Observations of a possible Gobi dust event by infrared lidar near Mauna Loa, Hawaii  
**M.J. Post and D.A. Bowdle**

Aerosol backscattering climatologies as observed by infrared lidar  
**M.J. Post**

An interactive model for predicting scattering functions of polydisperse aerosols  
**P.C. Reist and W. Wilson**

Concentration of some aerosol components and its influence on global solar radiation absorption and scattering in central part of upper Silasian Industry Region, Poland  
A. Sztyler

SESSION 4: TRANSPORT, SCAVENGING AND DEPOSITION

Background air quality and deposition of trace elements in the province of South-Holland  
J. van Daalen

Study of atmospheric aerosols in a terrain-induced nocturnal boundary layer using bistatic lidar  
P.C.S. Devara and P.E. Raj

Vertical profile and elemental concentrations of aerosols in a forest area  
K. Hameri, M. Kulmala, S. Ylatalo, U. Tapper and T. Raunemaa

Modelling dry deposition of particles to the ocean  
P. Hummelshoj and C. Hansen

Modelling of the transport of water-soluble aerosols by a tropical squall line  
M. Laube and P. Scheidgen

Transport, chemistry and deposition of airborne gaseous and particulate pollutants in a marine environment  
S.M. Joffre and V. Lindfors

Ionic balances in marine rainwater with a special emphasis on sources of alkaline and acidic species  
R. Losno, G. Bergametti, P. Carlier and G. Mouvier

Concentration profiles of trace elements over the North Sea and the Atlantic Ocean : anthropogenic and natural contribution  
R. Losno, P. Carlier and G. Bergametti

Diurnal variation of the concentrations of radon and its shortlived daughters in the atmosphere near the ground  
J. Porstendorfer, G. Butterweck and A. Reineking

Removal of airborne particles by raindrops  
M. Taberi and Sh. Ayatollahi

1800	Bus to hotels
1930	Bus departure to dinner
2000	Conference dinner at the Connemara Coast Hotel

(cont'd p 6) Transport, Sourcing and deposition - 1

Thursday 15 June

SESSION 4: TRANSPORT AND DISPERSAL OF AEROSOLS, vertical and horizontal.

0900 Long range transport of aerosols : invited review  
R. Jaenicke (e)(c)

0945 AEROSOL TRANSPORT (VERTICAL)

0945 Troposphere aerosol size distributions in eastern and northern North America between 1982 and 1988 : A signature for troposphere-stratosphere exchange?  
W.R. Leaitch and G.A. Isaac

1000 Aerosol measurements in the Southwest Pacific  
I.S. Lechner, G.W. Fisher, M.J. Harvey, H.R. Larsen and R.A. Knobben

1015 Vertical profiles of aerosol properties on the Arctic summer troposphere  
J. Heintzenberg, J. Strom, J.A. Ogren and L. Gardneus

1030 Vertical profiles of aerosol to 7 km in the arid Southwestern US  
R.G. Pinnick, G. Fernandez, D.M. Garvey and E.H. Holt

1045 Refreshment break

1115 AEROSOL TRANSPORT (HORIZONTAL)

1115 Atmospheric transport of sulfur and nitrogen across the North Atlantic ocean from North America to Europe  
J.N. Galloway and D.M. Whelpdale

1130 Long-range transport of trace metals to North Europe marginal seas  
G. Petersen, H. Weber, H. Grassl and J. Pacyna

1145 Source, transport and deposition of lead in the Mediterranean atmosphere  
E. Remoudaki, R. Losno, G. Bergametti and P. Buat-Ménard

1200 European source area identification and apportionment of long range transported aerosol  
E. Swietlicki, B. Swantesson and H.-C. Hansson

1215 Influence of ammonia from agriculture on trends in background aerosol concentrations  
H.M. ApSimon, M. Kruse-Plass, P. Stott and G. Whitecombe

1230 Atmospheric electricity in Galway  
J.C.H. van der Hage

1245 Lunch

1400                   **AEROSOL DISPERSAL - SCAVENGING**

1400                   Scavenging of aerosol particles by drizzle and precipitation sized drops  
                         B. T. McGann and S.G. Jennings

1415                   A theoretical study of the wet removal of two different types of aerosol particles by a two dimensional dynamic model for a convective warm cloud  
                         A.I. Flossmann and H.R. Pruppacher

1430                   A field study on in-cloud removal of aerosol particles  
                         T. Schumann

1445                   A theoretical investigation of the collection of aerosol particles by falling ice crystals  
                         N. L. Miller

1500                   The scavenging of high altitude aerosol by small ice crystals  
                         D.A. Bell and C.P.R. Saunders

1515                   Refreshment break

1545                   Modelling of wet scavenging of heavy metals in the marine environment  
                         L. Levkov, D.P. Eppel and H. Grassl

1600                   Field measurements of the wet deposition of particulate materials  
                         K. W. Nicholson and J.R. Branson

1615                   Sequential sampling of major ions, dissolved and total trace metals in wet deposition  
                         B. Lim and T. D. Jickells

1630                   Daily measurements of background air pollutants at a site in the west of Northern Ireland  
                         J. Stedman

1645                   Seasonal variations in sulfate, nitrate and chloride in the Greenland ice sheet : relation to atmospheric concentrations  
                         C.I. Davidson

1700                   Conference Close

## AEROSOL CLIMATOLOGY

O. Preining  
Institute for Experimental Physics  
University of Vienna  
Austria

Climatology in general became, during the past few years, a very hot issue. The global heating due to the greenhouse effect, the seemingly unavoidable global temperature increase caused by human activities, and the ozone hole, developing over the antarctic set an alarm. From the climate modelling efforts and from analyzing the situating and extrapolating into the next hundred years it became evident that only large scale changes in human activities by all mankind can reduce the problems of the future to manageable sizes. However these problems are only to a minor part scientific, they can not be solved by science but only by political decisions, very hard and difficult decisions, and the decision makers need a strong and convincing scientific backing to induce the general and global acceptance of the necessary restrictions.

In this situation the aerosol science community found itself asked to contribute. A very fast look around gave the astonishing fact that: 1) aerosols may play an important role for the assessment of climate, 2) there is little known about the atmospheric aerosol, its production, its residence time in the atmosphere, its sinks and its influences on climate. A joint working group (JWG) on an international aerosol climatology project (IACP) formed and issued a plan outline for a "Global Aerosol Climatology and Effects Program (GACEP)". In their summary they stated on the role of aerosols: Large fractions of the atmospheric aerosols are of biological origin and may have considerable influence on climate by acting as cloud and ice nuclei, especially sulfate particles forming in the atmosphere from dimethyl sulfide from marine organisms may also act as cloud nuclei. Aerosols may add to the greenhouse effect about as much as  $\text{CO}_2$ . The Ozone-hole is influenced and probably enforced (or even caused) by polar stratospheric clouds which in turn form on aerosol precursors, and finally Cloud Chemistry also depends on aerosols.

### Literature

Landolt-Bornstein, New Series  
Group V: Geophysics and Space Research, Vol.4, Meteorology  
Subvolume b: Physical and Chemical Properties of the Air,  
Springer 1988.  
Subvolume c: Part I, Climatology Part 1, Springer 1987  
Subvolume c: Part II, Climatology Part 2, Springer 1988

A Deepak and G. Vali ed's for JWG  
Global Aerosol Climatology and Effects Program (GACEP): A Plan  
Outline; June 1988.

## DEVELOPMENT OF A GLOBAL-SCALE BACKGROUND AEROSOL MODEL

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University of Alabama at Huntsville  
Huntsville, Alabama 35899  
USA

A global-scale model of background tropospheric aerosols is being developed to provide critical baseline design parameters for NASA's prospective space-based Laser Atmospheric Wind Sounder (LAWS). Precise definitions are required to properly design, communicate, and evaluate this research. Three general background classes are distinguished. The term operational background refers to the dominant modal structure in a distribution of aerosol measurements from a given aerosol sensor. Full specification of this term requires: 1) the measured or derived aerosol property; 2) the end-to-end measurement system, including aerosol sensor, sensor platform, data processing, and analysis; 3) the end-to-end accuracy, precision and detection threshold; 4) the volume, shape, position, orientation, and spatial/temporal resolution of the effective sample zone; 5) the spatial/temporal domain of the full data set; 6) the end-to-end sample selection criteria for the full data set (including operational constraints); 7) the methodology for identifying and isolating the background mode from the full data set; and 8) the statistical descriptors for the background mode. The term consensus background refers to a set of estimates of a given aerosol property, derived from operational background values for a wide variety of aerosol data sets, and determined to be similar by a given statistical test. Specification requires: 1) the data set selection criteria; 2) the data conversion methods; and 3) the statistical test for "similarity". A geophysical background refers to a set of airmasses with similar values of a given aerosol property, as deduced from the presence of a consensus background for the property. Application of this methodology to a specific problem requires careful choice of the full set of background parameters are sought from each data set in NASA's GLOBal Backscatter Experiment (GLOBE) data base on background tropospheric aerosols. This comprehensive global-scale data base includes measurements of aerosol microphysics and short wavelength aerosol optical properties, along with modelling and direct measurements of aerosol backscatter properties at CO<sub>2</sub> wavelengths. A global-scale consensus background of  $10^{-10} \text{ m}^2 \text{ kg}^{-1} \text{ sr}^{-1}$  was found for backscatter mixing ratios (aerosol backscatter coefficients normalized by air density) at the 9.11 micron design wavelength for LAWS, suggestive of a global-scale geophysical background for this property and a corresponding operational background design baseline for LAWS. Model methodology and results should be useful in: 1) studies of the life-cycles, global-scale budgets, and physicochemical properties of background aerosols; 2) the design of sensors that use scattering from background aerosols in the measurement of other atmospheric quantities; and 3) the development of data compression algorithms for massive global-scale aerosol data bases.

ARCTIC HAZE: LONG RANGE POLLUTION OF THE POLAR ATMOSPHERE

R.C. Schnell  
CIRES, University of Colorado  
Boulder, CO 80309, USA

J.D. Kahl  
Geophysical Monitoring for Climatic Change  
National Oceanic and Atmospheric Administration  
Boulder, CO 80303, USA

The Arctic Gas and Aerosol Sampling Program (AGASP) is a multifaceted cooperative research program designed to determine the distribution, transport, chemistry, aerosol physics, and radiative effects of the polar-wide air pollution phenomenon known as Arctic haze. The research was conceived, organized, and directed by the National Oceanic and Atmospheric Administration (NOAA), and the Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder. AGASP has involved participants from the United States, Canada, Norway, Sweden, Federal Republic of Germany, and Denmark and has covered two intensive field study periods in March-April 1983 and March-April 1986, and a third is scheduled for March-April 1989. The core field research program consists of airborne measurements tied to similar baseline station measurements at Pt. Barrow, Alaska; Alert, Northwest Territories; and Ny Alesund, Spitzbergen. The results of the 1983 program were published in special issues of Geophysical Research Letters (vol. 11, no.5, May 1984) and Atmospheric Environment (Vol.19, no.12, December 1985). The 1986 program results are "in press" in special issues of the Journal of Atmospheric Chemistry and Atmospheric Environment, both due out in mid-1989.

The program has shown that the winter-spring Arctic atmosphere is polluted with well-defined layers of anthropogenic gases and aerosols of Eurasian origin. Some of the pollution plumes have been tracked for over 10,000 km. The plumes contain excess amounts of C, CH<sub>4</sub>, CO<sub>2</sub>, CO, SO<sub>2</sub>, and a host of inorganic trace species.

Concern has been raised that this massive air pollution in the Arctic could be affecting climate by trapping radiation as haze optical depths of 0.75 have been measured. The haze also may be depositing toxic elements and acidic species on the ice caps and on the high Arctic tundra.

## SOUTHERN OCEAN CONDENSATION NUCLEI AND CLIMATE VARIATIONS

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Atmospheric condensation nucleus concentrations have been determined on a regular basis at Cape Grim, Tasmania since 1977. However, a clear understanding of what represents a "baseline" measurement has been elusive. The idea that a "baseline" for CN (and CCN) concentrations exists only in a statistical sense is proposed. Use of the concentration-frequency distribution to describe the aerosol is suggested, with the distribution geometric mean a single convenient measure. This approach has been followed with the Cape Grim CN data obtained from 1977 to 1987 to derive monthly estimates of "baseline" concentration. These show strong seasonal variations but also quite marked variations on an interannual time scale. By numerically filtering the monthly data the presence of quasi-periodic oscillations in "baseline" CN concentrations with frequencies similar to those observed in the Southern Oscillation have been revealed. Some association between the major fluctuations and El-Nino Southern Oscillation perturbations in the regional circulation is suggested and possible mechanisms discussed.

CLOUD CONDENSATION AND AITKEN NUCLEI MEASUREMENTS IN  
BACKGROUND AIR

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A series of measurements is described which was made at the Cape Grim Low Background Station in 1985 in which concentrations of CCN and Aitken nuclei were determined. The samples taken when background air was being sampled could be determined from the weather situation. In addition however it was possible to distinguish background samples not only from the lower concentration of particles but also from the shape of the curves of CCN concentration vs supersaturation.

AEROSOL CLIMATOLOGY MEASUREMENTS WITH NOLAN-POLLAK COUNTERS

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Galway, Ireland

The Nolan-Pollak counter was originally developed in the 1940's for aerosol climatology measurements although its versatility led subsequently to many other applications in aerosol science. It has been widely used in manual, automatic and modified forms in many parts of the world to record ambient concentrations of Aitken nuclei. Measurements at Mace Head, Ireland will be used to discuss its suitability for the purpose. The design, calibration and performance of an automatic counter will be discussed.

Acknowledgement: This work was supported by an Eolas Research Grant.

## MARINE AEROSOLS

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### INTRODUCTION

Knowledge of the physical and chemical properties of marine aerosol particles is important because of the role these particles play in a number of important atmospheric processes. Marine aerosols affect radiative transfer and climate directly by scattering and absorbing radiation and indirectly by influencing the droplet size distribution and albedo of marine boundary layer clouds. In addition, marine aerosol particles play an important role in the marine portion of the atmospheric sulfur cycle; they affect the performance of electro-optical systems since they limit transmission of electromagnetic radiation; and they provide the backscatter signal necessary to permit retrieval of wind profiles over the ocean using doppler-lidar wind sounder techniques (Gerber, 1989).

The number, size distribution and composition of marine aerosol particles vary in time and space in response to a number of different processes including long-range transport of continental air; aerosol production by the bursting of whitecap bubbles and by homogeneous nucleation involving the oxidation products of precursor gases emitted by the ocean; removal of particles by deposition and precipitation scavenging; and aerosol transformation processes such as coagulation, condensation of low volatility reaction products (e.g., sulfuric acid), and enhancement of aerosol mass as a result of  $\text{SO}_2$ -to-sulfate conversion in cloud droplets.

In the last fifteen years a number of excellent review papers on marine aerosols have been published. Junge (1972) presented a remarkably discerning survey of our knowledge of the physico-chemistry of aerosols in the undisturbed marine environment. On the basis of available data Junge developed an idealized model of the various components of the undisturbed marine aerosol. More recently, Podzimek (1980, 1982) reviewed the advances in marine aerosol research in the decade following Junge's review. Since 1980, considerable gains have been made in our knowledge of the marine aerosol, particularly with regard to the size distribution of submicron aerosols, the concentrations of the main aerosol constituents in clean marine air, and the processes which control the submicron portion of the size distribution.

The purpose of this review is to present an up-to-date description of our knowledge of the physico-chemical properties of aerosols in clean marine air, with emphasis on the following topics: the total particle concentrations; the size and volume distributions of particles over the size range  $0.006 < r < 50 \mu\text{m}$ ; the principal aerosol constituents and their concentrations; sources of the marine aerosol; and the dependence of the particle size distribution and salt loading on wind speed.

### TOTAL PARTICLE CONCENTRATIONS

Podzimek (1980), updating an earlier study of Hogan (1976), has summarized the results of numerous observations of Aitken nucleus

concentrations over the ocean in the form of a map showing the geographical distribution of mean Aitken nucleus concentrations over the oceans. Concentrations range from  $4000-6000 \text{ cm}^{-3}$  off the east coasts of North America and Asia to less than  $300 \text{ cm}^{-3}$  over the central regions of the Atlantic, Pacific and Indian Oceans. Hoppel et al. (1989) measured the variation in total particle concentrations (obtained by integrating the particle size distribution between  $0.006$  and  $0.5 \mu\text{m}$  radius) during a transatlantic cruise from Charleston, SC to the Canary Islands. Particle concentrations decreased from  $6000 \text{ cm}^{-3}$  near the coast to a mean of about  $200 \text{ cm}^{-3}$  over the remote Atlantic in air which had a long history ( $> 10$  days) over water. The lowest total particle concentration observed was  $100 \text{ cm}^{-3}$ . Particles smaller than  $0.03 \mu\text{m}$  radius contributed less than 20% to the total particle concentration in clean marine air over the subtropical North Atlantic. No correlation was found between total particle concentrations and wind speed.

Recent measurements of total particle concentrations have also been made in remote regions of the eastern Pacific Ocean. Parungo et al. (1987), using a GE automatic condensation nucleus counter having good sensitivity as low as  $10 \text{ cm}^{-3}$ , report mean concentrations of about  $125 \text{ cm}^{-3}$  along the Equator at  $5^\circ\text{S}$  when winds were from the Southern Hemisphere. The lowest concentration was  $60 \text{ cm}^{-3}$ . Hoppel and Frick (1989) found the mean concentration of particles larger than  $0.006 \mu\text{m}$  radius to be  $200 \text{ cm}^{-3}$  between Hawaii and Tahiti on a cruise from Hawaii to the South Pacific. The lowest average daily total particle concentration  $70 \text{ cm}^{-3}$  measured near  $55^\circ\text{S}$  and  $155^\circ\text{W}$ . Precipitation was reported the previous day.

Diurnal variations in total particle concentrations over the open oceans have been reported by several investigators. Meszaros and Vissy (1974) found a noontime maximum in the average diurnal change in the concentration of Aitken particles over the South Atlantic and Indian Oceans. A diurnal variation in Aitken nucleus concentrations, with a daytime maximum and nighttime minimum, was observed over the Pacific by Parungo et al. (1987) only when the air mass was constant throughout the day. A daytime maximum in Aitken particle concentrations implies photochemical production of very small particles which may be due to higher daytime photooxidation rates of organosulfur gases (such as DMS) emitted by the ocean. However, Hoppel et al. (1989) and Hoppel and Frick (1989) found no evidence of a daytime maximum in the concentration of particles larger than  $0.006 \mu\text{m}$  over the remote Atlantic and Pacific Oceans. This finding may not be inconsistent with a diurnal variation in Aitken concentrations if the daytime maximum is due to variations in the concentration of particles smaller than  $0.006 \mu\text{m}$ . One would expect, however, that, once produced, such small particles would grow to  $0.006 \mu\text{m}$  and larger. Haaf and Jaenicke (1980) did not find a diurnal variation in total particle concentrations over the North Atlantic but did find a pronounced noon maximum in the concentration of particles smaller than  $0.01 \mu\text{m}$  radius. Bigg et al. (1984) found seasonal variations in Aitken particle concentrations with maximum at the summer solstice and minimum in winter at five remote Southern Hemisphere sites. The variations, which ranged from an order of magnitude difference at Syowa ( $69^\circ\text{S}, 40^\circ\text{E}$ ) to a 25% difference at Samoa ( $14^\circ\text{S}, 171^\circ\text{W}$ ), were attributed to variations in the photochemical production of particles related to the availability of solar radiation.

#### PARTICLE SIZE DISTRIBUTION

Figure 1 is a plot of the aerosol particle size distributions in clean marine air, as measured by various investigators in the last decade. Table 1 summarizes the location, sampling height and method of measurement for each of the distributions. The distributions of Exton et al. (1986), Hoppel et al.

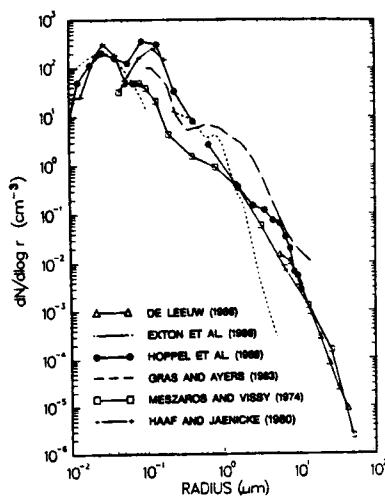


Fig. 1. Published measurements of the size distribution of aerosol particles in clean marine air.

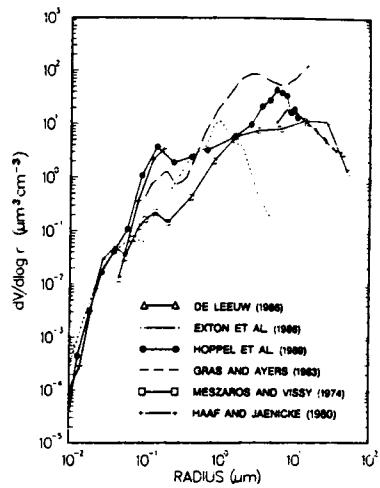


Fig. 2. Particle volume distributions corresponding to the size distributions in Figure 1.

(1989) and de Leeuw (1986) were obtained at wind speeds of  $5 \text{ m s}^{-1}$  or less, while the spectrum of Meszaros and Vissy (1974) is a mean over a period when the average wind speed was  $12 \text{ m s}^{-1}$ . The distribution of Gras and Ayers (1980) is a geometric mean of measurements made over a period of several months during which the wind speed ranged from  $3 \text{ m s}^{-1}$  to  $20 \text{ m s}^{-1}$ . Data on the size distribution of particles larger than  $10 \mu\text{m}$  radius are particularly scarce. It is difficult to determine to what extent the differences in the size distributions are due to differences in sampling location, to differences in wind speed (in the case of particles larger than  $0.3 \mu\text{m}$  radius), or to

TABLE 1. SUMMARY OF THE SAMPLING LOCATION, SAMPLING HEIGHT AND METHOD OF MEASUREMENT FOR THE PARTICLE SIZE AND VOLUME DISTRIBUTIONS DISCUSSED IN THE TEXT.

AUTHORS	LOCATION	SAMPLING HEIGHT (m)	INSTRUMENTATION AND PARTICLE SIZE RANGE (μm)
MESZAROS AND VISSY (1974)	SOUTH ATLANTIC AND INDIAN OCEANS ( $40^{\circ}$ - $68^{\circ}$ S)	SHIP LEVEL	OPTICAL AND ELECTRON MICROSCOPE ANALYSIS OF FILTER SAMPLES ( $0.02 < r < 60 \mu\text{m}$ )
GRAS AND AYERS (1980)	CAPE GRIM, TASMANIA ( $40^{\circ}$ S, $144^{\circ}$ E)	98	DIFFUSION BATTERY WITH POLLAK CNC ( $0.002 < r < 0.1$ ) CLIMET OPC ( $0.15 < r < 5.0$ )
HAAF AND JAENICKE (1980)	NORTH ATLANTIC DURING JASIN ( $89^{\circ}$ N, $10^{\circ}$ W)	14	ELECTRIC MOBILITY ANALYZER ( $0.003 < r < 0.4$ )
DE LEEUW (1986)	NORTH ATLANTIC ( $87^{\circ}$ N, $20^{\circ}$ W)	11	ROTORDR INERTIAL IMPACTOR ( $8.5 < r < 42.5$ )
EXTON ET AL. (1986)	COASTAL SITE ON OUTER HEBRIDES ( $67^{\circ}$ N, $7.5^{\circ}$ W)	10	PMS ASAS AND CSA PROBES ( $0.08 < r < 10$ )
HOPPEL ET AL. (1986)	SUBTROPICAL ATLANTIC ( $20$ - $30^{\circ}$ N, $15$ - $70^{\circ}$ W)	SHIP LEVEL	DIFFERENTIAL MOBILITY ANALYZER ( $0.008 < r < 0.6$ ) REPLICATOR IMPACTOR ( $0.6 < r < 16$ )

differences in the method of measurement. The low concentrations of large particles measured at Cape Grim, Tasmania, may be due in part to the higher sampling height (95 m), but studies of the vertical distribution of sea-salt particles over the ocean (Blanchard et al., 1984; Warneck, 1988) indicate less than a factor of three change in concentration between 15 and 100 m.

An important feature exhibited by all of the size spectra is a decrease in slope, or flattening, in the range of 0.2 - 0.5  $\mu\text{m}$  radius. This may be a consequence of the fact that this size range is a transition region between size ranges having totally different sources of particles.

The development of the differential mobility analyzer (Hoppel, 1978; Haaf, 1980a; 1980b) resulted in more accurate measurements of the submicron aerosol size distribution than was possible with previous methods such as counting of filter samples, diffusion batteries and electrical precipitation (Jaenicke, 1980). Differential mobility analyzer measurements show that the submicron aerosol size distribution in clean marine air over the remote oceans is bimodal with one peak in the range of 0.02-0.03  $\mu\text{m}$ , the other in the range of 0.09-0.15  $\mu\text{m}$  (Haaf and Jaenicke, 1980; Hoppel et al., 1986; Hoppel et al., 1989). Hoppel et al. (1985, 1986) examined a number of mechanisms which might produce a bimodal submicron aerosol size distribution. It was concluded that the cycling of the aerosol through cloud formation and evaporation processes (i.e., non-precipitating cloud cycles) was the mechanism most likely responsible for the formation of the double peak. During the cloud phase, trace gases (e.g.,  $\text{SO}_2$ ) are absorbed by cloud droplets and are converted to particulate matter such as sulfates. Consequently, the nuclei remaining after the cloud droplets evaporate are larger than the original particles on which the droplets formed and this results in a separation in the size distribution between those particles which serve as cloud condensation nuclei and those which are too small to be active.

Although the names attached to different modes in the size distribution is somewhat arbitrary, the mode at 0.1 - 0.15  $\mu\text{m}$  can appropriately be referred to as the accumulation mode since particles of this size fall in the size range for which the total time constant for removal due to all processes is a maximum (Hoppel et al., 1989). Precipitation scavenging is the most efficient mechanism for removing particles from this size range.

As pointed out by Gras and Ayers (1983), the few reported measurements made in the difficult to measure size range of  $r < 0.005 \mu\text{m}$  show a wide variance in particle concentrations. Jaenicke (1979), using a diffusion battery method, found considerable concentrations of such small particles with a peak around 0.002  $\mu\text{m}$  radius, in clean air at Tasmania. Such results indicate continuous production of particles by gas-to-particle conversion. However, diffusion battery measurements made by Gras and Ayers (1983) at the same location under similar clean conditions do not show a persistent mode at 0.002  $\mu\text{m}$ .

#### PARTICLE VOLUME DISTRIBUTION

Figure 2 shows the particle volume distributions calculated from the size distributions in Figure 1. While there are considerable differences in the volume distribution at radii greater than several microns including differences in the location of the large particle peak, all of the distributions are characterized by two distinct modes with a minimum in the range of 0.2 - 0.3  $\mu\text{m}$  radius. The two modes are referred to as the fine (small) particle mode and the coarse (large) particle mode. The coarse mode constitutes about 90-95% of the total mass but only 5-10% of the total number

of particles. The coarse particle mass in clean marine air is comprised mainly of sea salt and shows a strong dependence on wind speed.

#### COMPOSITION AND SOURCES OF THE MARINE AEROSOL

The principal constituents of the marine aerosol are sea salt, non-sea-salt sulfate, mineral dust, and, generally to a lesser extent, nitrates. Non-sea-salt sulfates and nitrates have both a continental and marine source. Mineral dust is transported from semi-arid and desert regions and is a highly variable component of the coarse particle mode. The concentration of sea salt depends primarily on wind speed and can range from about  $2 \mu\text{g m}^{-3}$  to over  $100 \mu\text{g m}^{-3}$ . The primary marine sources of  $\text{NO}_x$  and hence nitrate are downmixing from the stratosphere and production by lightning (Crutzen and Gidel, 1983). Table 2 gives the concentrations of the aerosol constituents in clean marine air over the remote oceans, as measured by various investigators. Although sea salt does not have a continental source and does not, strictly speaking, have a clean marine or background concentration, it is included in Table 2 for

TABLE 2 CONCENTRATIONS ( $\mu\text{g/m}^3$ ) OF THE PRINCIPAL AEROSOL CONSTITUENTS IN CLEAN MARINE AIR.

CONSTITUENT	MASS CONCENTRATION	LOCATION	REFERENCE
SEA SALT	2.6 - 8.0	EQUATORIAL AND TROPICAL PACIFIC GALAPAGOS TO HAWAII	RAEDONCK ET AL. (1986)
	2.7 - 38.0	N. ATLANTIC ( $60^\circ\text{N}$ , $10^\circ\text{W}$ ) $U < 10 \text{ m/s}$	LOVETT (1978)
	1.2 - 68.0	S. ATLANTIC ( $40-80^\circ\text{S}$ , $40-88^\circ\text{W}$ ) $U < 10 \text{ m/s}$	LEPLEE ET AL. (1983)
	2.0 - 20.0	CAPE GRIM, TASMANIA $U < 10 \text{ m/s}$	GRAS AND AYERS (1983)
NON-SEA-SALT SULFATE	0.3 - 1.1	EAST EQUATORIAL PACIFIC ( $8^\circ\text{S}$ , $110-140^\circ\text{W}$ )	PARUNGO ET AL. (1987)
	0.2 - 0.85	EASTERN TROPICAL PACIFIC	RAEDONCK ET AL. (1986)
	0.18 <sup>a</sup>	PANAMA TO TAHITI	WAENHAUT ET AL. (1983)
	0.38	INDIAN OCEAN CLEAN S. HEMISPHERE AIR	SAVOIE ET AL. (1987)
MINERAL DUST	0.1	EQUATORIAL AND TROPICAL PACIFIC GALAPAGOS TO HAWAII	RAEDONCK ET AL. (1986)
	0.07	GALAPAGOS TO TAHITI	WAENHAUT ET AL. (1983)
	0.2	PACIFIC MARINE BOUNDARY LAYER ( $12^\circ\text{N}$ TO $48^\circ\text{S}$ )	PATTERSON ET AL. (1980)
	0.27	INDIAN OCEAN, S. HEMISPHERE AIR	SAVOIE ET AL. (1987)
	0.04 - 0.45 <sup>b</sup>	CLEAN NORTH PACIFIC AIR	UEMATSU ET AL. (1983)
	0.08 - 0.1	TROPICAL CENTRAL PACIFIC	UEMATSU ET AL. (1983)
	0.01 - 8.0 0.38 <sup>c</sup>	CENTRAL AND NORTHERN N. ATLANTIC	PROSPERO (1979)
NITRATE	0.16	INDIAN OCEAN, S. HEMISPHERE AIR	SAVOIE ET AL. (1987)
	0.1 - 0.2	EAST EQUATORIAL PACIFIC	PARUNGO ET AL. (1987)

<sup>a</sup> SINGLE VALUES ARE ARITHMETIC MEANS EXCEPT AS NOTED

<sup>b</sup> RANGE OF MEAN VALUES AT SEVERAL ISLAND LOCATIONS

<sup>c</sup> GEOMETRIC MEAN

the purpose of comparing the relative contribution of the various constituents to the total aerosol mass in clean marine air. The concentrations of sea salt given are for wind speeds less than  $10 \text{ m s}^{-1}$ . Sea salt concentrations can be as high as  $100 \mu\text{g m}^{-3}$  at wind speeds in excess of  $20 \text{ m s}^{-1}$  (Exton et al., 1986; Lovett, 1978).

#### Non-sea-salt Sulfate

The concentration of non-sea-salt sulfate decreases markedly from coastal regions of the continents to the remote areas of the oceans. Parungo et al. (1987) found that non-sea-salt sulfate concentrations were almost an order of magnitude lower over the equatorial Pacific than they were off the west coast of the United States. As reported by Parungo et al. (1986, 1987), non-sea-salt sulfate concentrations over the remote oceans have been found to increase in regions of ocean upwelling and can at times exceed the concentrations of sea-salt sulfate.

Numerous studies have shown that the fine particle mode ( $r < 0.3 \mu\text{m}$ ) in clean marine air is composed predominantly of non-sea-salt sulfate. Examination of individual particles by means of electron microscopy (Meszaros and Vissny, 1974; Gras and Ayers, 1983; Parungo et al., 1987) has shown that a high percentage of the fine-mode particles are morphologically similar to ammonium sulfate. Small amounts of sulfuric acid have also been detected.

Aerosol volatility measurements have also shown that the majority of small particles are composed of non-sea-salt sulfate. Clarke et al. (1987) measured the change in the particle size distribution in clean Pacific air and found that only about 1% of the fine-mode particles survived heating to temperatures that would volatilize everything but sea salt. Approximately 40% of the particles disappeared at temperatures that would volatilize sulfuric acid particles, the remaining 60% having a thermal response like ammonium sulfate/bisulfate. Hoppel and Frick (1989) also measured the volatility of submicron particles ( $r < 0.5 \mu\text{m}$  radius) over the remote Pacific Ocean and found that, typically, only a few percent of the particles were involatile. However, measurements made on a day with very clean air (total particle count of  $70 \text{ cm}^{-3}$ ), following a couple of days of stormy conditions, showed that 40% of the particles smaller than  $0.2 \mu\text{m}$  radius were involatile and presumably composed of sea salt. Thus, it appears that enough small salt particles can be produced during high winds to make a significant contribution to the fine-particle mode in very clean background air.

A question of major importance is the origin of the non-sea-salt sulfate in clean marine air and the relative contributions from long-range transport of continental air and in situ production over the ocean. The evidence for a significant marine source of non-sea-salt sulfate is substantial and includes the following: (1) the presence of methanesulfonic acid (which has no continental source) in marine aerosol particles, (2) higher concentrations of non-sea-salt sulfate in ocean upwelling regions, (3) a sufficient source strength of reduced sulfur gases from the ocean to sustain the observed concentrations of excess sulfate against removal processes, (4) an apparent source of small particles which maintains the small particle concentrations against coagulation decay and (5) the existence of a double peak in the submicron aerosol size distribution.

The photooxidation of dimethyl sulfide (DMS) gas, which is produced by biological activity in the ocean and released into the atmosphere, is believed to be the most important source of marine background concentrations of non-sea-salt sulfate (Andreas and Raemdonck, 1983; Cline and Bates, 1983; Bigg et al., 1984). In the atmosphere, DMS is rapidly oxidized by the OH radical to  $\text{SO}_2$  and methanesulfonic acid (MSA), the proportion of  $\text{SO}_2$  being as much as 80%

at the low  $\text{NO}_x$  levels typical of the remote marine atmosphere (Tyndall et al., 1986).  $\text{SO}_2$ , in turn, is photooxidized to  $\text{H}_2\text{SO}_4$  by reaction with OH and other free radicals (Eggleton and Cox, 1978).

Sulfate is formed by gas-to-particle conversion involving the oxidation products of DMS. The principal gas-to-particle conversion mechanisms are: (1) Formation of new particles by heteromolecular homogeneous nucleation in the  $\text{H}_2\text{SO}_4$ -MSA-water system, with possible involvement of other trace gases such as ammonia (2) Condensation of  $\text{H}_2\text{SO}_4$  and MSA on newly-formed and existing particles and (3) Aqueous-phase oxidation of  $\text{SO}_2$  to sulfate in cloud droplets. All three of these processes influence the submicron aerosol size distribution over the remote oceans (Fitzgerald and Hoppel, 1988; Hoppel, 1988).

The relative importance of the different gas-to-particle conversion pathways as a source of the non-sea-salt sulfate in marine aerosols is not fully resolved. Predicted gas-phase  $\text{H}_2\text{SO}_4$  concentrations equal to only 10% of the background sulfates (Toon et al., 1987, Kritz, 1983) and MSA to non-sea-salt sulfate ratios less than 0.1 (Saltzman et al., 1983, 1985) tend to argue against homogeneous oxidation of  $\text{SO}_2$  to  $\text{H}_2\text{SO}_4$  and condensation of MSA and  $\text{H}_2\text{SO}_4$  as main pathways from DMS to aerosol sulfate. Rather,  $\text{SO}_2$ -to-sulfate conversion in clouds may be the most important pathway. It should be noted, however, that concentrations of MSA and  $\text{H}_2\text{SO}_4$  in marine aerosols may be low because of reaction of  $\text{H}_2\text{SO}_4$  with ammonia to form ammonium sulfate and by oxidation of MSA by OH in aerosols and cloud droplets (Saltzman et al., 1986). It is further noted that significant amounts of  $\text{H}_2\text{SO}_4$  in marine aerosols have been reported by Clarke et al. (1987).

#### Sea Salt

Sea salt is the principal constituent of the coarse-particle mode except during episodes of continental dust transport. The production of sea-salt aerosol is due to the agitation of the sea surface by the wind. Wind stress on the ocean surface is a generator of air bubbles which later burst, producing both film drops and jet drops (Blanchard and Woodcock, 1957; Blanchard, 1963). Bubbles are most numerous in the whitecaps associated with breaking waves, where they are formed by the entrainment of air into the surface water by breaking wave motion. Whitecaps begin to appear at wind speeds of about  $3 \text{ m s}^{-1}$ . At wind speeds of  $10 \text{ m s}^{-1}$ , approximately 1% of the sea is covered by whitecaps and at  $15 \text{ m s}^{-1}$  4% of the surface is covered (Monahan, 1971; Toba and Chaen, 1973). Each bursting bubble produces between 1 and 10 jet drops and up to several hundred smaller film drops, depending on the size of the bubble. After generation, the drops rapidly equilibrate to the ambient relative humidity and become either sea-salt particles or sea-salt solution droplets. Most film and jet drops have radii  $< 10 \mu\text{m}$  and most film drops are smaller than  $1 \mu\text{m}$  radius (Woolf et al., 1988).

In addition to indirect production of sea-salt particles by bursting bubbles, there is also direct production of spume drops resulting from the mechanical disruption of wave crests by the wind. Spume drop formation becomes a significant contributor of large ( $> 10 \mu\text{m}$  radius) spray droplets at wind speeds in excess of  $10 \text{ m s}^{-1}$  (Wang and Street, 1978; Monahan et al., 1986). Spume drop formation has been invoked to explain the 'tail' on the large droplet end of aerosol spectra measured during the 1978 JASIN Experiment (Monahan et al., 1983).

Figure 3 shows the dependence of sea-salt aerosol mass concentration on wind speed, as reported by several investigators. Each curve is a linear least squares fit of the expression  $\ln \theta = b + aU$ , where  $\theta$  is the salt mass concentration and  $U$  is wind speed, to scatter plots of sea-salt mass versus wind speed. Sea-salt concentrations are determined either by computing the

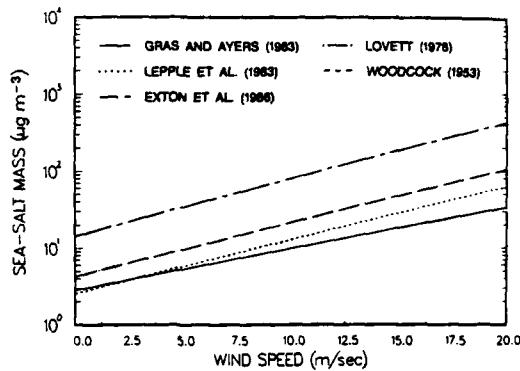


Fig. 3. Sea-salt mass concentration as a function of wind speed.

integrated aerosol volume from measurements of the particle size distribution or by measuring the sodium content of the salt solution extracted from aerosol samples. The individual scatter plots show that salt mass concentrations can vary by as an order of magnitude or more at a given wind speed. Factors contributing to this scatter include variations in wind speed history of the air mass (i.e., whether wind speeds are increasing or decreasing), variations in the stability structure of the marine boundary layer, variations in sea surface temperature which may affect viscosity and bubble production, and scavenging effects of precipitation.

Monahan and colleagues (Monahan et al., 1982, 1986) have used measurements of aerosol production in the University College, Galway, whitecap simulation tank to develop a model of the open-ocean production rate of sea-spray droplets per unit area of sea surface, per increment of droplet radius, as a function of 10-m wind speed. Such a model is an important element in the development of a time-dependent model of the aerosol size distribution in the marine boundary layer.

#### Mineral Dust

Mineral dust is a highly variable constituent of the marine aerosol. It is transported to the marine atmosphere from semi-arid and desert regions when wind patterns are favorable. The coarse mode almost always contains a mineral dust component, but mean dust concentrations over most ocean areas including the North Atlantic, the tropical Pacific and the Southern Hemisphere oceans are typically less than  $0.5 \mu\text{g m}^{-3}$  and can be as low as  $0.05 \mu\text{g m}^{-3}$  (Prospero, 1979; Raemdonck et al., 1986; Savoie et al. 1987). However, high dust concentrations, sometimes exceeding the sea-salt concentration, can occur over certain ocean regions such as the tropical and equatorial North Atlantic Ocean, the northwest Indian Ocean and the Peru/Ecuador shelf when atmospheric wind patterns transport dust from the desert areas of northern Africa, the Arabian Peninsula and the coastal areas of Peru and Ecuador (Raemdonck et al., 1987, Prospero, 1979). Elevated dust concentrations can also be found over the North Pacific Ocean during certain times of the year (Uematsu et al., 1983). Prospero (1979) found that dust concentrations over the tropical and

equatorial North Atlantic can vary by a factor of 100 depending on the origin of the air.

In clean, remote ocean areas, 70% of the mass of mineral dust is associated with particles larger than 1  $\mu\text{m}$  radius (Raemdonck et al., 1986) and a large fraction of the dust component is found to be internally mixed with sea-salt particles (Raemdonck et al., 1986; Andreae et al., 1986). Andreae et al. (1986) suggest that the internal mixing could be produced by processes in clouds, including coagulation of droplets formed on the two types of particles and/or the scavenging of the crustal particles by cloud droplets.

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## MARINE AEROSOLS GENERATED FROM BURSTING AIR BUBBLES

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Marine aerosols are generated by bursting air bubbles at the sea surface; those produced through the fragmentation of film cap are film drops, and those through the break up of a water jet formed by the collapse of bubble cavity are jet drops (Blanchard 1963). These drops are associated with the cloud formation, air-sea exchanges, sea-surface enrichment, as well as ocean-engineering practices. Consequently, there have been many attempts in quantifying productions of these drops. The observation on film drops, because of their vastness in number and smallness in size, is especially difficult. Two successful experiments, however, were conducted recently over the still water surface with artificially produced bubbles (Resch et al. 1986; Blanchard and Syzdek 1988). Their results are herewith reviewed and found to be highly complimentary; Resch et al. covered the largest size possible down to about  $8 \mu\text{m}$  in diameter, and Blanchard and Syzdek extended from there to say  $0.01 \mu\text{m}$ . Excluding the peculiar production peak observed in the latter, both sets of data are found to follow very closely the same power law; their combination provides the total production function:  $n_f = 1.07 D^{2.15}$  where  $n_f$  is the number of film drops produced by their parent bubble having the diameter  $D$  (mm). The production of jet drops was studied earlier by Kientzler et al. (1954) and summarized by Blanchard (1983). Their data on the number of jet drops produced by parent bubbles were parameterized by Wu (1988b) as:  $n_j = 7 \exp(-D/3)$ . In the meantime, the bubble concentration at

the sea surface was proposed by Wu (1988a) as:  $N_0 = 57 U_{10}^{3.5} D^{-4}$ , where  $U_{10}$

expressed in  $\text{ms}^{-1}$  is the wind velocity measured at 10 m above the mean sea surface and  $D$  is again expressed in mm. Combining all these results, the production of marine aerosols under various wind velocities are estimated; they consist mainly of jet drops.

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SUMMERTIME AEROSOL MEASUREMENTS IN THE ROSS SEA REGION  
OF ANTARCTICA

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Measurements of atmospheric aerosols have been made in the Ross Dependency, Antarctica in the 1986-87 and 1988-89 summer field seasons. The study is part of a N.Z. Meteorological Service programme which aims to characterize the physics and chemistry of atmospheric particulate matter in the S.W. Pacific at sites remote from anthropogenic influences.

The results presented will show size distributions 0.1 - 3.0  $\mu\text{m}$  dia determined by a single particle optical counter, C.N. concentrations and chemical composition determined by both X-ray emission analysis of individual particles (TEM/EDAX) and bulk filtration. Data collected during a three week study on Bowers Piedmont Glacier in November 1988 showed particle concentration (0.1 - 3.0  $\mu\text{m}$  dia) to range between 25 - 50  $\text{ml}^{-1}$  and distributions to be bimodal (with large particle mode of about 1  $\mu\text{m}$  dia). Similar distributions of particles were observed at three other sites in 1986-87. Initial examination by EDAX shows most sub- $\mu\text{m}$  diameter particles to be sulphur species.

Some comparisons will be made between this data, collected at the surface and measurements made in the free troposphere between 1.5  $^{\circ}\text{N}$  (Tarawa) and 46  $^{\circ}\text{S}$  (Invercargill, N.Z.) from an aircraft which has been instrumented by N.Z. Met. Service.

## PHYSICAL CHARACTERISTICS OF THE AMBIENT AEROSOL AT MACE HEAD

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Measurements of the ambient aerosol number and particle size distribution were made at Mace Head, on the west coast of Ireland, to obtain the physical characteristics of particles in the maritime air from the North Atlantic ocean and in airmasses from the continent of Europe. The aerosol particle sizing instrumentation, based on optical scattering counters, had a continuous measurement capability over a radius range of 0.045 to 1.5 micrometres with automated data handling down to 2 Hz. This was supplemented by manual and automatic Nolan-Pollak Aitken nuclei counters. Field measurements from late 1987 to date are presented on particle number, size, mass and surface distributions.

The measurements show a reduction of between two or three orders in magnitude of these aerosol parameters when the wind is from a W or SW direction (that is for a maritime air mass) as compared to when the aerosol is of continental European origin from a predominantly E or SE direction. Curve fitting of the particle size data for the prevailing SW airmass at Mace Head indicated that in general there were three log-normal modes present. The smaller mode possessed geometric mean radius  $r_g$  of between 0.07-0.08  $\mu\text{m}$  and geometric standard deviation  $\sigma_g$  of about 1.30; the second mode possessed  $r_g$  and  $\sigma_g$  values of about 0.17  $\mu\text{m}$  and 1.1 whilst the third mode was characterized by a geometric mean radius of about 0.3  $\mu\text{m}$  and geometric standard deviation of about 1.9. Aerosol mass loadings varied from  $10.5 \mu\text{g m}^{-3}$  (for W direction air trajectory) up to  $48.7 \mu\text{g m}^{-3}$  for the E - SE wind sector of anthropogenic source. The mass measurements were complemented by Aitken nucleus measurements at the site which showed that the lowest particle number concentration counts (less than  $100 \text{ cm}^{-3}$ ) originated from the W direction whilst the highest particle concentration (greater than  $10^4 \text{ cm}^{-3}$ ) were from the SE direction.

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NORTH ATLANTIC AEROSOL  
BACKGROUND CONCENTRATIONS MEASURED AT A HEBRIDIAN COASTAL SITE

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At the beginning of a series of field observations in the Outer Hebrides during August 1986, there was a period of several days when the wind speed remained below 7m/s, which contrasted dramatically with the earlier Spring observations with wind speeds gusting to 50 m/s. During this calm period, there was little or no whitecapping, beyond the surf zone, and very low particle counts were recorded for all measured sizes (radii from 0.09 to 23.5  $\mu\text{m}$ ). These data provide estimates of the very low wind speed background particle counts characteristic of the North Atlantic environment and, also, yield information on particle loss rates. The background count previously obtained by extrapolating the concentration/wind speed relationship to zero wind speed was considerably higher; this was probably due to the short times spent at lower wind speeds being insufficient to permit the particle concentration to reach its equilibrium level.

When periods of heavy precipitation and continental influence are removed from these data, there was generally a steady decline in aerosol concentration throughout this episode for particles from about 0.25 to 16  $\mu\text{m}$  radius. The concentration of the largest particles,  $16 < r < 23.5 \mu\text{m}$ , was almost uniformly zero throughout, with only the occasional particle recorded. For smaller particles, no discernible trend could be established suggesting either very low loss rates or, more likely, continuing production from sources other than the sea surface.

Mechanisms of particle loss including sedimentation, coagulation and dry deposition were considered and the observed time constants were compared with estimates derived from simple assessments of these processes. An elementary model of particle loss by dry deposition to the water surface is adequate to explain the observed time constants and, for most particle sizes turbulent deposition dominates gravitational sedimentation.

PHYSICAL AND CHEMICAL PARAMETERS OF MARINE BACKGROUND  
AND SAHARA AEROSOLS OVER THE NORTH ATLANTIC OCEAN

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During the last 20 years, several aerosol measuring campaigns have been carried out on board of research vessels in the Eastern North Atlantic and during field expeditions at various West African locations. The samples were taken at different seasons of the year. Number and mass concentrations as well as size distributions have been determined with various aerosol sampling and measuring systems. Two major aerosol bodies are likely because there are moderate variations of the concentration levels for major parts of the aerosol size distribution, and a similar shape of size distribution as well as a rather constant relative elemental composition. Therefore, marine background and Sahara aerosols can be described with typical distribution and concentration parameters.

MEASUREMENTS OF AEROSOL CONCENTRATION AND DISTRIBUTION AT  
HELGOLAND ISLAND

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Environmental aerosols cover a size range from a few nanometers up to several micrometers. For particle sizing of such aerosols different sensors based on different principles of operation have to be applied. In the present study a Differential-Mobility-Particle-Sizer (DMPS), a laser aerosol spectrometer model LAS-X and a white light optical particle counter model DAP-Test-2000 have been combined. The DMPS, which classifies particles according to their mobility, is applied for measurements between 10 and about 200 nm. The LAS-X which usually delivers an optical diameter, can be used in the size range between 90 nm and about 3  $\mu$ m and the DAP-Test-2000 for particles between 1  $\mu$ m and 25  $\mu$ m. Since the optical diameter depends on the refractive index of the particles, direct calibrations of the LAS-X in terms of mobility-diameters have been carried out (Brand and Gebhart, 1988). Because of its high time resolution (5 min), this instrument allows observations of short-term variations in particle number, volume and size distribution. In general the aerosol number concentration was nearly one order of magnitude lower than in urban districts, but particle production periods were observed in which the concentration was comparable to values measured in cities. These production periods were correlated to sun radiation and the generated particles were found in a nucleation mode in the diameter range between 20 nm and 40 nm.

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## VOLATILITY OF BACKGROUND AEROSOL

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In this work the volatile properties of Atlantic background maritime aerosol is reported for the first time. Field measurements on the volatility of background aerosol particles have been carried out at Mace Head field research station on a remote location off the west coast of Ireland, spanning 18 months from December 1987 to date.

Evaporation in aerosol particle size occurred as the particles are drawn through a quartz heated tube whose inner temperature was varied from ambient temperature up to 850 °C. Detailed measurements of the reduction in particle size and number concentration with temperature were made generally over 60 minute temperature cycles with a Particle Measurement Systems (PMS) light-scattering aerosol counter - an Active Scattering Spectrometer Probe (ASASP-X).

The field volatility measurements show a sharp decrease in aerosol number concentration for particle radius up to about 0.4 micrometers for temperatures up to 400 °C.

A comparison of the field data was made with laboratory aerosol polydispersion volatility measurements for a wide range of atmospheric aerosol constituents using the identical volatility apparatus. Average field data temperature fractionation curves for submicrometre particles compare favourably with the laboratory data (up to 400 °C) for ammonium sulphate (and ammonium bisulphate) suggesting that those materials are a major constituent of the submicrometre marine background aerosol fraction.

Field volatility measurement for higher temperature up to 850 °C show that the particle size radius range (0.3 - 1.5 micrometre) is also volatile and display temperature - fractionation characteristics of sodium chloride, on comparison with laboratory data. The measurements also indicate that the majority of the remaining submicrometre aerosol is largely unaffected at these higher temperatures suggesting the presence of clay mineral type constituents in the background submicrometre aerosol.

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INTERHEMISPHERIC VARIABILITY IN AEROSOL PHYSICOCHEMISTRY  
OVER THE REMOTE PACIFIC OCEAN

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Extensive measurements of aerosol physicochemistry and related properties have been carried out over the remote Pacific using research vessels, aircraft and a stationary site at Mauna Loa Observatory, Hawaii (alt. 3,500 m). A custom 256 channel optical particle counter matched via microcomputer to a thermal conditioning system acted as a Thermo-Optic Aerosol Discriminator (TOAD). This sulfuric acid, sulfate or sea-salt to be separately accumulated in-situ and in real-time. Coincident measurements of condensation nuclei (CN), aerosol light scattering coefficient (integrating nephelometer), aerosol light-absorption (integrating sandwich photometer) were also made.

Data from the North Pacific generally show elevated coarse and fine mode aerosol concentrations at higher latitudes than the equatorial and southern Pacific. The total CN and the relative fraction of CN remaining after heating to 300 C also appear greater in the northern hemisphere and often when light-absorption values are highest, usually indicating the presence of soot. The ratio of ammonium to sulfate in the aerosol, as inferred by the TOAD system, is also higher in the northern hemisphere (usually near one or greater) than in the southern hemisphere (usually near zero). These observations suggest the North Pacific is often influenced by continental and combustion aerosol with higher ammonia concentrations than the southern hemisphere. The relative "cleanliness" of the south Pacific atmosphere with a  $\text{NH}_4^+/\text{SO}_4^{2-}$  value that tends to be zero, except when air masses indicate recent passage over land, suggests that natural sources of sulfuric acid are common to extensive oceanic regions. Experiments using a differential mobility analyzer/TOAD combination indicate an internally mixed fine-mode aerosol in the North Pacific but a small externally mixed component south of the equator.

COMPARISON OF AEROSOL CHARACTERISTICS IN THE ARID SOUTHWESTERN  
US AND THE WEST COAST OF WESTERN EUROPE

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The addition of a heated intake to a particle counter, as employed in the pioneering work of Rosen (1) and Twomey (2), permits the volatility of the ambient aerosol to be determined, leading to information about the composition of the aerosol particles. This technique has been used for aerosol measurements in two diverse locations - the west coast of Western Europe and an interior basin in the arid southwestern United States. A common bimodal aerosol distribution is found at both sites. The smaller fraction of the aerosol is found to be dominated by ammonium sulphate in both cases. A dependence of aerosol number density on air mass type is also a common feature. In the southwestern US the change from a modified maritime polar air mass to a modified maritime tropical air mass by a frontal passage led to a concentration increase of up to an order of magnitude in the volatile submicron particles and a substantial increase in the proportion of nonvolatile particles in this size range. On the west coast of Ireland aerosol number density increases by two to three orders of magnitude as the air mass changes from maritime to continental origin. The nonvolatile aerosol fraction in the southwestern US is found to be composed of quartz and clay minerals with some carbonaceous particles suspected in low concentration. The use of higher instrument intake temperatures for analyzing the aerosol on the west coast of Ireland led to the identification of sodium chloride in the supermicron aerosol fraction. Planned installation of aethalometers at both locations is designed to determine the carbonaceous content of the aerosol and its dependence on air mass origin and meteorological condition.

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DISTRIBUTION OF METHANESULFONATE, NSS-SULFATE AND  
DIMETHYLSULFIDE OVER THE ATLANTIC AND THE NORTH SEA

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The emission of gaseous biogenic sulfur compounds from marine and continental sources is an important supplier of atmospheric sulfur. One of its essential processes is the flux of dimethyl-sulfide (DMS) from the oceans to the atmosphere with an estimated amount of 40 Tg S(DMS)/yr, about half of the global anthropogenic sulfur emissions. In the atmosphere DMS is oxidized by OH-, NO<sub>3</sub>- and IO<sub>3</sub>- radicals. Final oxidation products are sulfate (SO<sub>4</sub><sup>2-</sup>) - and methanesulfonate (CH<sub>3</sub>SH<sub>3</sub><sup>-</sup>, MSA) - particles which are effective as condensation nuclei. Information about the yield of these two compounds are still discrepant.

The results of parallel measurements of methanesulfonate and non-sea-salt (nss)-sulfate in the aerosol and of dimethylsulfide in the marine atmosphere are presented with the main emphasis on the MSA-aerosol. These investigations were performed during two passages with the research vessel "Polarstern" across the Atlantic between 50°N and 30°S in March/April 1987, September/October 1988 and during measurements on the island Sylt (May 1986, August 1987) as well as on a research platform in the North Sea (May 1988). In addition the concentrations of MSA and nss-sulfate were determined in rain- and fog-water.

The concentration of MSA in the aerosol varied between 1 and 20 ng S(MSA)/m<sup>3</sup> over the Atlantic while in the region of the North Sea higher values of 5 - 100 ng S(MSA)/m<sup>3</sup> were observed. On an average the S(DMS)-concentration was a factor of 4-9 higher than the S(MSA) - concentrations. Beyond the influence of continents the quotient of nss-sulfate- and MSA-concentrations in the aerosol was about 10. The MSA-concentration proved to be a good indicator for advection of marine air masses. During the measurements on the platform a diurnal variation of the MSA- concentration was found with a maximum at daytime. In rain-water MSA-concentrations of 1 - 20 µg S(MSA)/l were determined, while in fog the values increased to 50- > 100 µg S(MSA)/l. The concentration of MSA showed a reverse correlation with the pH of rain- and fog-water.

## THE CHEMISTRY OF AEROSOLS IN THE ATMOSPHERE

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The chemical composition of atmospheric aerosols is influenced by three major factors:

- (a) composition of primary aerosol emissions of both natural and man-made substances;
- (b) composition of secondary aerosols formed by gas to particle conversion processes;
- (c) subsequent chemical reactions of the primary and secondary aerosols.

Typical example of aerosol components arising by all three mechanisms will be presented.

There are many techniques by which chemical information upon atmospheric aerosol composition may be obtained. If it is required to characterize the aerosol beyond the level of elemental abundance in a bulk sample, two techniques stand out as especially valuable:

- (a) X-ray powder diffraction, which can provide an unequivocal identification of specific crystalline chemical compounds within a size-fractionated bulk sample,
- (b) electron microscope or microprobe techniques in which the elemental composition of individual particles is determined.

Some of the results of the application of such methods will be reviewed, and the conclusion presented that irrespective of geographic location, there are major similarities in aerosol composition worldwide.

Two aerosol systems providing a particular chemical challenge are the  $\text{NH}_4\text{NO}_3$  /  $\text{NH}_3$  /  $\text{HNO}_3$  and  $\text{NH}_4\text{Cl}/\text{NH}_3/\text{HCl}$  systems. These are described and discussed in the light of the ability of equilibrium chemical thermodynamics to predict their status, and of experimental studies designed to elucidate the kinetics of achievement of equilibrium conditions.

ELEMENTAL COMPOSITION OF AEROSOL PARTICLES UNDER  
BACKGROUND CONDITIONS IN HUNGARY

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The elemental composition of atmospheric aerosol particles sampled under regional background conditions is determined by using the PIXE method. The results of a sampling program of several years are presented. It is shown that sulfur and silicon are the most abounding elements showing that the aerosol is partly of anthropogenic and partly of soil origin. The origin of particles is further discussed on the basis of elemental ratios related to selenium and vanadium as well as on the basis of the analyses of meteorological conditions during sampling. The results are compared to those obtained under oceanic conditions to estimate the strength of continental sources. Finally, for some cases the regional and long range transport of different elements is evaluated.

PARTICULATE AND GASEOUS AIR POLLUTANT LEVELS  
AT THE PORTUGUESE WEST COAST

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A field study has been conducted on a coastal site near Aveiro-Portugal to assess the chemical composition of aerosol particles and the levels of inorganic gaseous compounds involved on gas/particle conversion in the atmosphere. For this purpose parallel sampling of suspended particles with a "high volume" sampler and HCl, HNO<sub>3</sub> and NH<sub>3</sub> with a filter pack technique was carried out during 24 hour periods. The aerosol samples were analysed for sulphates, nitrates, chlorides, ammonium, organic carbon, elemental carbon and trace elements including K, Na, Mg, Ca, Be, Cd, Cr, Cu, Fe, Mn, Mo, Ni, Pb, V and Zn by a variety of techniques (atomic absorption, atomic emission, ion chromatography, thermal volatilization/NDIR, etc.)

Gaseous and particulate pollutant levels are similar to other values observed in non-polluted atmospheres. The equilibrium between particle and gas phase is evaluated for NH<sub>4</sub>Cl and NH<sub>4</sub>NO<sub>3</sub> compounds in comparison with thermodynamic data.

Backward air mass trajectories and source apportionment techniques were applied to the data to find the groups and processes responsible for the aerosol burden in this region of Europe.

## ACID AEROSOL MEASUREMENTS AT A SUBURBAN CONNECTICUT SITE

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Atmospheric acidity data was gathered during a year-long field project investigating the possible health effects of acid aerosols at a rural site in southwestern Connecticut. This site was chosen as the air quality is frequently influenced by pollutants transported from the New York City-New Jersey corridor.

An annular denuder filter-pack system was utilized to obtain daily measurements of gaseous  $\text{HNO}_3$ ,  $\text{HONO}$ ,  $\text{SO}_2$  and  $\text{NH}_3$ , fine particle  $\text{SO}_4^{=}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and  $\text{H}^+$ , and fine mass ( $<2.1 \mu\text{m}$ ). Ozone concentrations and basic meteorological data were also obtained continuously. Multiple measurement locations, in a 10 mile radius of the central site, were also utilized to study the spatial patterns of aerosol acidity in the community. Preliminary data from the summer of 1988 at this site will be presented and contrasted to data taken at other network sites. Correlations of atmospheric acidity with ozone and meteorological parameters will be presented.

SENSITIVITY LIMIT, RESOLUTION, COUNTING EFFICIENCY  
AND COINCIDENCE LIMIT OF OPTICAL AEROSOL PARTICLE COUNTERS

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The importance of aerosol contamination monitoring in the semiconductor industry is well established and recognized to achieve high yields in Integrated Circuit production. The trend of this industry to smaller dimensions forces instrument manufacturers to push the performance of optical aerosol particle counters (OAPC) to their physical limit. The driving force is the desire to detect the smallest particle. Unfortunately lowering the sensitivity limit of an OAPC might also affect other important instrument performance parameters.

This paper will illustrate the relationship between four major OAPC performance parameters and will expose their individual effect on instrument performance. It will be shown that many instruments with low sensitivity limits ( $0.1 \mu\text{m}$  and lower) can only accomplish this task if counting efficiency is sacrificed to less than 50% and resolution is reduced. In reality this means that instrument performance is reduced in order to detect "some" of the small particles which then define the sensitivity limit of the instrument.

CHARACTERIZATION OF THE CARBONACEOUS AEROSOL IN THE WESTERN  
MEDITERRANEAN ATMOSPHERE

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Aerosols were sampled over the Western Mediterranean sea during 4 field experiments and analysed for their total and soot carbon contents. Particulate carbon is always the major element of the aerosol. It exhibits rather uniform atmospheric concentrations (mean values: 2 and 0.4  $\mu\text{gC}/\text{m}^3$  for total and soot carbon respectively). The two components display a satisfactory covariation which suggests a common origin. The carbonaceous aerosol bears the characteristics of that sampled in industrial source regions: on the average, 22% of the carbon is in the soot mode and analyses of size-segregated samples indicate that at least 50% of the carbon mass is attached to submicron-particles. These results suggest that the particulate carbon produced from fossil fuels is conservative during its transport in the atmosphere. Also, intense anthropogenic atmospheric inputs in Western Europe generate an important background of carbonaceous particles over the Mediterranean sea. Such an aerosol load might influence the transport and chemistry of other compounds as well as the radiative balance of this atmospheric environment.

INTERNAL/EXTERNAL MIXING OF SOOT TO AEROSOLS IN SOURCE  
AND RECEPTOR REGIONS IN AUSTRIA

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Elemental carbon (soot) can be mixed to the atmospheric aerosol either internally (i.e. within the individual particles) or externally. The type of mixing determines the optical properties of the aerosol and especially its single scattering albedo, which is often used to predict the radiative and climatic consequences caused by an aerosol layer. Model calculations (e.g. Ackerman, Toon 1981) have shown that, depending on the mixing type, a certain amount of elemental carbon can produce non-negligible differences of the single scattering albedo of a model aerosol.

During an intensive measurement period of two weeks in September 1983 the mass size distribution of the aerosol in the urban source area of Vienna and in a rural receptor region in its vicinity was measured with AERAS cascade impactors. The impaction foils were subsequently analyzed for total mass, sulfate, nitrate and carbon. A summary of these measurements was presented elsewhere (Hitzenberger et al. 1985), while the humidity dependence of the size distributions of the chemical species is investigated in the present paper. If the aerosol is mixed externally, the size of the (nonhygroscopic) elemental carbon particles should not be a function of relative humidity. If, on the other hand, elemental carbon is present within the hygroscopic salt particles, the carbon size distribution should undergo the usual humidity dependent growth/evaporation processes.

From the chemical size distributions in Vienna no appreciable change of the carbon distribution was found for relative humidities above 50%. The shape of the distribution stayed more or less constant during the whole measurement period. The sulfate and nitrate distributions follow the expected humidity pattern, so one could conclude that at least in the source region the aerosol is mixed mainly externally. In the same time period, however, all measured chemical size distributions in the receptor region exhibit similar humidity trends. The carbon size distribution closely follows that of the salts. Therefore an internal mixture seems to be indicated.

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A METHODOLOGY FOR ESTIMATING REGIONAL EMISSIONS OF BLACK  
CARBON IN THE ARCTIC

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Black Carbon aerosol (BC), a strong absorber of visible light, constitutes a small but significant fraction of the effluent arising from carbonaceous fuel combustion processes. Estimates of the source strengths and geographical distribution of BC emissions, needed to model their effects on radiative transfer, are currently unavailable. In this paper we present a methodology for estimating regional emissions of BC in Arctic latitudes based on the synthesis of three independent pieces of information: time-resolved measurements of BC and CO<sub>2</sub> at Pt. Barrow, Alaska; long-range atmospheric trajectory analyses; and tabulated values of regional fuel consumption. Preliminary results imply annual BC emissions in the Novaya Zemlya/Taymyr Peninsula regions of the northern USSR on the order of 10<sup>8</sup> g yr<sup>-1</sup>.

AIRBORNE PARTICULATE ELEMENTAL CARBON :  
ITS SOURCES, TRANSPORT AND SOILING POTENTIAL

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This paper will review the sources of airborne particulate elemental carbon (PEC) and develop an emission inventory for this pollutant. This inventory shows that diesel emissions are the main source of PEC, even for sites remote from highways. Observed correlations between PEC and Pb are in good agreement with their respective emission factors.

The application of a model for area ground level emissions is considered. The predictions of the model are consistent with observed values.

Correlations between gravimetric and smoke shade estimations of airborne particulate mass are considered and the relative contribution from PEC assessed. The contribution of PEC to airborne dark smoke varies from 60% in urban areas to a national average of 25%. Finally, the soiling potential of particulate matter deposited from the atmosphere to horizontal and vertical surfaces, and the relative contribution from PEC, will be assessed and compared with experimentally obtained values.

#### PARTICLE SAMPLES FROM FOREST FIRES

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Naturally occurring forest fires are one of the contributors to the background atmospheric aerosol. The general perception has been that forest fires will give rise to sooty and carbonaceous smoke and the sampling of particles from them is of some interest especially vis a vis nuclear winter studies. It is not generally possible to obtain close in samples of the aerosol from natural forest fires, but in Ontario forest management includes prescribed burns in which areas of unproductive forest are set alight prior to replanting.

In 1987 a helicopter was used to obtain particle samples fairly close to a prescribed burn near Timmins, Ontario. Particles were sampled qualitatively using a rather crude but effective 'whirling broomstick' technique. It was found that sooty carbonaceous particles were the exception rather than the rule and the samples revealed, rather surprisingly, a wide variety of particle types depending on the closeness of the sampling point to the fire and the time after formation. Some hypotheses will be advanced, rather tentatively, in an attempt to account for these differences.

## ATMOSPHERIC AEROSOL SOURCES AND THEIR CLIMATIC SIGNIFICANCE

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Specifications of the sources and emission rates of atmospheric particulate matter provide basic information for estimating their climatic impact both on a regional and global scale. The production sources and mechanisms reflect the chemical composition, the size range, and the shape of the particles. These properties indicate the ability and the extent to which the particles can interact with electromagnetic waves, liquid and solid matters and to go or not into solution droplets.

In the present study, a survey of the following species: sulphate, sea-salt, soot, mineral, dust-like, and water-soluble substances have been made to determine their microphysical properties and their radiative characteristics as a function of the ambient relative humidity. Their global distributions and their variations with time have been made to evaluate their climatic signature in the visible and infrared regions. Updated results of the study will be presented.

ATMOSPHERIC OPTICAL DEPTHS: BACKGROUND AND PERTURBATIONS  
IN THE TROPOSPHERE AND STRATOSPHERE

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Sun photometry is a convenient way to measure atmospheric optical depths, one of the parameters by which climatically important aerosol-related perturbations of the background atmosphere can be assessed. Traditionally, sun photometric observations have been ground-based. Such observations do not provide information on the vertical profile of optical depths, thereby limiting an assessment of climatic effects of background and pollution aerosols. This situation has been changed in recent years with the advent of airborne sun photometry (Matsumoto et al., 1987); Russell et al., 1986; Pueschel et al., 1988) and spaceborne extinction measurements (McCormick, 1986; Kent and McCormick, 1984). In this presentation we compare surface-derived atmospheric optical depth data from WMO's Background Air Pollution Monitoring Network (BAPMON) with airborne results from NASA's First ISLCP (International Satellite Land Climatology Project) Field Experiment (FIRE), and with extinction measurements of NASA's Stratospheric Aerosol and Gas Experiment (SAGE) satellite program. This data evaluation permits a distinction between tropospheric and stratospheric optical depths, and their perturbations by anthropogenic and volcanic pollutants, respectively.

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SPECTRAL EXTINCTION COEFFICIENTS OF BACKGROUND AEROSOLS IN  
EUROPE, NORTH AMERICA AND SOUTH AMERICA; A COMPARISON

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Several intensive studies have been performed in order to investigate the atmospheric aerosol in remote locations and to obtain data on the atmospheric extinction coefficient. The following results were obtained:

A) Europe: In an early study in the late summer of 1964 the size distribution was measured continuously with an aerosol spectrometer. The number size distribution functions had a peak around 0.3 micrometer, secondary maxima sometimes occurred above one micrometer diameter. The extinction coefficient of the aerosol was calculated by means of Mie theory, and values between 0.03 and 0.2 km have been found. The lowest values occurred during snowfall, high values normally were obtained when the haze layer extended to the elevation of the measuring site, which was at an altitude of 2000 meters.

Similar results have been obtained from determinations of the spectral extinction coefficient with the university of Vienna telephotometer, which were performed since 1972. On days with excellent (European) visibility the extinction coefficient usually has a value of 0.03 km, on average days values around 0.1 km are typical. It is interesting to note, that these values were found both in the Alps and at a rural site 100 km south of the heavy industrialized Ruhr district in Germany. This indicates, that the aerosol found at remote sites in Europe apparently is the average pollution spread over a large area.

B) North America: Spectral extinction coefficients were obtained with the University of Vienna telephotometer at a rural site north of Denver, Colorado, and at a site in the desert of northern Arizona in the Summer of 1979. Usually the extinction coefficients had values between 0.01 km and 0.04 km. At the rural site in Colorado an influence of the urban plume of Denver was found, since in most cases the values for sight paths towards the town were lowest, whereas in the desert site in the southwest the same extinction coefficient was measured in all directions. A severe smog episode in Los Angeles (more than 700 km to the west) caused an increase of the extinction coefficient by a factor of 2 at this remote location.

C) South America: In the Cordillera De Los Andes near Santiago de Chile extinction coefficients have been determined at elevations above 3000 meters. Values between 0.018 km and 0.15 km have been found depending on the height of the haze layer, which usually increases from below 2000 meters in the morning to above 4000 meters in the afternoon. Probably the low extinction coefficients in the morning still are influenced by "left over" aerosol of the haze layer of the previous day. This will be clarified by measurements, which are scheduled after the deadline for submission of abstracts.

EVIDENCE OF A TROPOSPHERIC AEROSOL BACKSCATTER BACKGROUND  
MODE AT CO<sub>2</sub> WAVELENGTHS

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The Laser Atmospheric Wind Sounder (LAWS) has been proposed for the measurement of global tropospheric winds. LAWS, a satellite-borne, lidar-based sensor, will measure Doppler shifts in aerosol backscatter signals. The wind measurement accuracy will depend critically on the strength of the backscattered signal, which in turn is a function of the backscatter coefficient  $\beta$  ( $\pi$ ), or  $\beta$ . During this decade, climatologies of backscatter profiles have been obtained in the troposphere and lower stratosphere at CO<sub>2</sub> wavelengths from several locations. Three such climatologies are described and compared. The Royal Signals and Radar Establishment (RSRE), United Kingdom (UK), obtained sounding measurements over the UK from 1981-1983 using the Laser True Airspeed System (LATAS), an airborne focussed 10.6  $\mu\text{m}$  pulsed Doppler lidar. The Jet Propulsion Laboratory (JPL) obtained measurements over Pasadena from 1983-1988 using ground-based 10.6 and 9.25  $\mu\text{m}$  lidars. The measured profiles were converted to backscatter mixing ratio ( $\beta/\rho$ ), a conserved quantity in the absence of source/sink processes, by normalizing the backscatter estimates ( $\text{m}^{-1}\text{sr}^{-1}$  units) by air density  $\rho$  ( $\text{kg m}^{-3}$  units) at each altitude. The resultant  $\beta/\rho$  profiles were averaged over selected height intervals and sorted into sub-decade classes of  $\log_{10}(\beta/\rho)$ . The frequency of occurrence corresponding to each bin at every height level was then calculated, color-coded, and displayed with computer-generated graphics for interpretation. Comparison of the resulting, "stacked" frequency histograms at 10.6  $\mu\text{m}$  from each location show striking similarities. A well-defined clean background mode ("background branch") within a narrow range of  $\beta/\rho$  values about  $10^{-10}\text{m}^2\text{kg}^{-1}\text{sr}^{-1}$  is apparent in the middle and upper troposphere. The extension of the background branch into the lower troposphere varies with the seasonal strength and geographic variability of a "convective branch" as determined by the convective boundary layer (CBL) activity and the surface aerosol flux. The position of the "stratospheric branch" varies with volcanic activity and the daily and seasonal rise and fall of the troposphere. The existence and similarity of the three background modes is remarkable in view of the natural aerosol backscatter variability (over four orders of magnitude in the troposphere), the diversity of the measurement conditions, and the presence

of stratospheric aerosol injections from Mount St. Helens (May 1980), El Chichon (Spring 1982), and other volcanoes preceding and throughout the measurement period. The 9.25 and 10.6  $\mu\text{m}$  JPL histograms are quite similar, except that the 9.25  $\mu\text{m}$  modal values are larger by approximately a factor of three above the CBL, consistent with theoretical predictions. It is noted that low aerosol extinction coefficients at 1.0  $\mu\text{m}$ , which could represent a background mode, have been observed in SAGE II solar occultation data. Empirical conversion factors between such a 1.0  $\mu\text{m}$  extinction background and the 10.6  $\mu\text{m}$  backscatter background are consistent with theoretical values. In summary, the existence of the background mode has major implications for global-scale aerosol models and studies of global aerosol budgets. It is also important for the design and performance simulation of prospective satellite sensors that rely on atmospheric aerosols as scattering targets.

THE MAUNA LOA AEROSOL BACKSCATTER INTERCOMPARISON  
EXPERIMENT (MABIE)

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Coordinated measurements of aerosol physicochemical and optical properties were obtained at the Mauna Loa Observatory (MLO) Hawaii, at 3.4 km altitude, during the Mauna Loa Aerosol Backscatter Intercomparison Experiment (MABIE) from November 14 - December 12, 1988. MABIE instrumentation included a high-volume filter/cascade impactor sampler (Georgia Institute of Technology); a high-resolution optical particle counter with thermal preconditioning (University of Hawaii); and a multiwavelength integrating nephelometer (NOAA's Geophysical Monitoring for Climatic Change) - all drawing from tall sampling stacks at the MLO site; and a high-power tunable (9.25  $\mu\text{m}$  or 10.6  $\mu\text{m}$ ) pulsed CO<sub>2</sub> Doppler lidar (NOAA's Wave Propagation Laboratory), situated at 3.2 km altitude, 2.9 km from MLO. Routine MABIE operations concentrated on concurrent measurements with all instruments in nocturnal downslope drainage flows, since these conditions supposedly transport background mid-tropospheric aerosols over the MLO sampling stacks. In addition, the lidar obtained several detailed vertical cross-sections of aerosol backscatter and winds in the upslope and downslope "skin" flows, along with numerous vertical profiles of backscatter and winds. Fair weather dominated MABIE sampling conditions, although brief stormy periods occurred during occasional frontal passages, with intervening periods of scattered maritime convection near the islands. Preliminary data during typical fair weather periods indicate background aerosol concentrations of approximately 0.1  $\mu\text{g m}^{-3}$ , with sulphuric acid apparently the dominant sub-micron component; scattering extinction at 0.55  $\mu\text{m}$  of approx.  $2-4 \times 10^{-7} \text{m}^{-1}$ ; and backscatter coefficients at 10.6  $\mu\text{m}$  of approx.  $10^{-11} \text{m}^{-1} \text{sr}^{-1}$ .

Background aerosol optical properties calculated from MABIE microphysical data are reasonably consistent with simultaneous measurements of those same optical properties. Moreover, measured and modelled optical properties from MABIE are reasonably consistent with background free tropospheric data obtained over less remote regions. These fair-weather MABIE data will be presented along with the rest of the MABIE data set in a local and regional meteorological context. This analysis is expected to determine whether downslope aerosol measurements at MLO are truly representative of the free troposphere above the island, and, consequently, representative of the free troposphere in the central Pacific Ocean around the islands. The results of this study will be used as inputs to a global-scale model of aerosol backscatter as part of NASA's GLObal Backscatter Experiment (GLOBE), IN SUPPORT OF NASA's prospective Doppler Laser Atmospheric Wind Sounder (LAWS).

## AEROSOL OPTICAL PROPERTIES IN REMOTE MARINE REGIONS

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During the past several years we have measured aerosol light-absorption,  $b_a$ , and aerosol light-scattering coefficients,  $b_s$ , in remote regions of the Atlantic, Pacific and Indian oceans. The light-absorption coefficient frequently reflects the concentration of soot carbon in the atmosphere and is a sensitive indicator of air masses influenced by combustion aerosol. This has enabled us to identify remote regions influenced by continental and/or anthropogenic influences and their optical properties.

Taken together the terms  $b_a$  and  $b_s$  define the single scatter albedo ( $SSA = b_s / (b_s + b_a)$ ), an important parameter for the determination of aerosol radiative effects in the atmosphere. Our measurements of  $b_a$  and the related soot concentrations range over 3 orders of magnitude for these remote regions. Lowest values were encountered in the South Pacific ( $b_s = 1 \times 10^{-8} \text{ m}^{-1}$ ) and highest values ( $b_s = 1500 \times 10^{-8} \text{ m}^{-1}$ ) were found over the South Atlantic. The related variability in SSA ranged from 0.99 to 0.77 respectively. The former value appears representative of "clean" marine regions with negligible continental influence and results in a net atmospheric "cooling" effect due to aerosol while the latter reflects continental combustion sources that result in a net atmospheric "heating". If our collection of data from these extensive oceanic regions is representative then SSA values representing "clean" conditions occur in only about half of the remote regions studied.

CONFERENCE POSTER PAPERS

PRESENTATION

## CONTINUOUS BACKGROUND AEROSOL MONITORING WITH THE EPIPHANIOMETER

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A new device was developed, which allows continuous aerosol monitoring. Calibration experiments showed that the device delivers a signal which is essentially proportional to the exposed aerosol surface, and it was therefore called epiphanometer (greek epiphanie = surface of a body). Its high sensitivity allows measurements also at very low concentrations found e.g. at high-alpine sites. Due to the low flow rate of 1 l/min the device consumes only 8 W and is therefore suitable for battery operation.

The system is based on a nuclear technique. Aerosols are pumped through a chamber containing radioactive lead isotopes ( $^{211}\text{Pb}$ ). These single lead atoms are produced at a constant rate by the decay of a short lived radon isotope emanating from a long-lived actinium source.  $^{211}\text{Pb}$  atoms attached to aerosols are transported through a capillary acting as a diffusion barrier for non-attached lead atoms. At the end of the capillary, the aerosols and with them attached lead atoms are deposited on a filter. The resulting activity on the filter is measured continuously by a surface barrier detector. Due to the relatively short half-life of  $^{211}\text{Pb}$ , the device allows continuous monitoring of aerosols, without changing or transporting the filter.

Results from a battery-operated aerosol monitoring station on Colle Gnifetti (Monte Rosa region, Switzerland, 4450 m a.s.l.) will be presented. The data are compared to two neighbouring stations at lower levels (3100 and 1600 m a.s.l.) and another high-alpine station (Jungfraujoch, 3450 m a.s.l.).

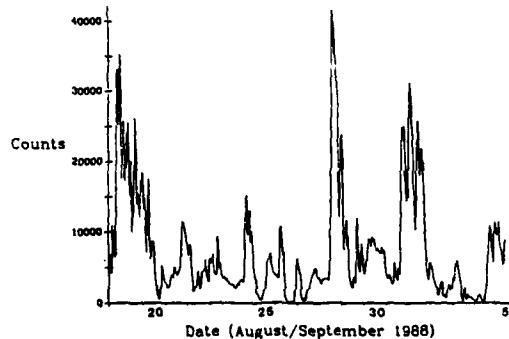


Figure 1. Signal of the epiphanometer for a 3 week measuring period on Colle Gnifetti, Monte Rosa (4450 m a.s.l.)

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H.W. Gaggeler, U. Baltensperger, M. Emmenegger, D.T. Jost, A Schmidt-Ott, P. Haller, M. Hofmann, "The epiphanometer, a new device for continuous aerosol monitoring", accepted for publication in J. Aerosol Sci.

A THERMO-OPTIC TECHNIQUE FOR IN-SITU ANALYSIS OF  
SIZE-RESOLVED AEROSOL PHYSICOCHEMISTRY

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We have developed a technique for the in-situ investigation of aerosol physicochemistry as a function of size over the range from  $0.14 \mu\text{m}$  to  $6 \mu\text{m}$ . The method is designed for use in remote background regions where the predominant fine-mode aerosol ( $D_p < 0.5 \mu\text{m}$ ) is generally sulfuric acid, ammonium sulfate/dust, sea-salt with some sulfate or nitrate). Interpretation of the data depends upon the thermal volatility of the various constituents and knowledge of their state of hydration and optical properties.

The optical components of a laser optical particle counter, OPC (LAS-X, Particle Measurement Systems, Boulder CO), have been used in conjunction with a custom 256 channel pulse height analyser (Norm Ahlgquist, University of Washington, Seattle, WA) installed in an Apple II microcomputer to create a high-resolution single range particle sizing instrument. The OPC is interfaced with a multichannel, continuous-flow heater assembly that is also under microcomputer control. The combined system acts as a real-time Thermo-Optic Aerosol Discriminator (TOAD). Aerosol is brought into the TOAD, mixed 1:1 with dry air (0% RH) and distributed to three heater tubes maintained at  $40^\circ\text{C}$ ,  $140^\circ\text{C}$  and  $300^\circ\text{C}$  after which it is selected and sized.

Analysis of the data is made by subtracting individual conditioned size distributions in order to isolate size distributions of the above mentioned components. These volume distributions are then "corrected" by relating volume ratios of volatiles lost at  $140^\circ\text{C}$  and  $300^\circ\text{C}$  to molar ratios of the inferred constituents via laboratory calibrations that compensate for refractive index and residual water. Quantitative agreement between results from standard analytical chemistry and TOAD inferred aerosol physicochemistry for both laboratory tests and several field studies will be demonstrated.

## BACKGROUND AEROSOL MEASUREMENTS IN GERMANY

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The atmospheric aerosol is a three phase system (solid particles with liquid shell in a carrier gas) and acts as a vehicle for the transport of materials of anthropogenic origin between sources and sinks. The particulate load of the atmosphere has changed in recent times from the dominant natural sources to anthropogenic ones. An increase of the load at smaller aerosol-particle radii has become evident besides the overall increasing output of pollutants from various sources. Changes in industrial processes like increased mass fluxes at higher production temperatures, production of synthetic materials from powder or gases, filter installations, waste removal and increasing traffic emissions caused a shift towards smaller radii in the distribution of primary particles released to the atmosphere. The small radii fraction is further augmented by the change in concentration and composition of gas emissions, because favourable meteorological conditions lead to the enhanced production of ultra fine secondary particles from the gas phase (under irradiation). This increase of air-borne particles in the small size range causes a substantially increased residence time of anthropogenic emissions. That again results in a transport of particulate matter over long distances into areas which formerly have been regarded as clean air regions. Only washout after cloud and precipitation scavenging efficiently reduces aerosol concentrations and contributes, besides other processes, to pollution deposition in the biosphere. Acidification of haze, fog and rain are measured and remobilisation of fairly toxic substances in the ground are observed.

The measurement system in use are a series of well designed low pressure impactors covering the size range from the nucleation to the coarse mode. Size selective measurements at 5 stations in Germany are compared according to their distribution and assorted chemical constituents.

COMPARISON OF AEROSOL SIZE DISTRIBUTION MEASUREMENTS  
IN AN URBAN AND REMOTE SITE IN SWITZERLAND

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The orography of the city Luzern is characterized by high altitude mountains in the South and East direction and low hills towards West and North. In the hilly region the major emission sources are situated and two highways entering the town centre. Between the mountains and the town a great lake provides ventilation. Vertical profiles and scavenging at the mountain slope has been published previously (Schumann et al, 1988). The measuring campaign has been concentrated on size selective particle measurements by low pressure impactors; the collection of meteorological data, on large and micro scale; traffic evaluations and the measurement of  $\text{NO}_x$ ,  $\text{SO}_2$ ,  $\text{CO}$ , TSP at two sites. The results prove the dominant role of particulate Nitrate as major pollutant in the town area. In consideration of the meteorological and automotive originated data the fluxes from the city towards the rural sites are calculated. The dispersion of the particulates, the gases and the production of particulates from the gas phase is the major input of the urban sites to the background aerosols at the remote sites. The industrial contribution to the background pollution are below the part of the traffic especially during rush hours. The very high number of particulates  $< 0.5 \mu\text{m}$  diameter increases the properties of CN and generates a stable layer of haze from the city towards the remote hill area at certain altitude.

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REACTION OF MONOTERPENES WITH OZONE, SULPHUR DIOXIDE  
AND NITROGEN DIOXIDE -GAS-PHASE OXIDATION OF SO<sub>2</sub>  
AND FORMATION OF SULPHURIC ACID

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Teflon bag experiments were carried out in the dark in order to study the gas-phase reactions of selected monoterpenes with ozone in the presence of SO<sub>2</sub> ( $\beta$ -pinene) as well as in the presence of SO<sub>2</sub>/NO<sub>2</sub> ( $\alpha$ -pinene,  $\beta$ -pinene, limonene). Emphasis was given in identifying the main reaction products and in quantifying the formed sulphuric acid aerosol. Apart from the sulphuric acid aerosol no other sulphur containing compounds could be detected.

It was found that the reaction of  $\beta$ -pinene with ozone, SO<sub>2</sub> and NO<sub>2</sub> leads mainly to 6,6-dimethyl-bicyclo [3.1.1] heptan-2-one (nopinone), the  $\alpha$ -pinene -O<sub>3</sub> -SO<sub>2</sub> -NO<sub>2</sub> - reaction produced 2', 2' - dimethyl -3-acetyl cyclobutyl ethanal (pinonaldehyde). The reaction of limonene with ozone -SO<sub>2</sub> -NO<sub>2</sub> leads mainly to an unidentified product with a molecular weight M = 134. In addition to the above mentioned volatile products, the formation of organic nitrates could be by means of gas chromatography - mass spectrometry established.

The yield of H<sub>2</sub>SO<sub>4</sub> in the system  $\beta$ -pinene /O<sub>3</sub> /SO<sub>2</sub> varies between 0.13 and 0.44 depending on the initial conditions e.g. humidity. In the system terpene/O<sub>3</sub> /SO<sub>2</sub> /NO<sub>2</sub> the yield of H<sub>2</sub>SO<sub>4</sub> was for  $\alpha$ -pinene, 0.01 - 0.03, for  $\beta$ -pinene 0.07 - 0.13 and for limonene 0.02 - 0.09.

## ATMOSPHERIC CONDITIONS OF PHOTOCHEMICAL SMOG FORMATION IN CITIES

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As it is known, anthropogenic impurities coming into the surface layer of the atmosphere of modern industrial cities undergo chemical reactions and form new compounds - secondary pollutants of the air. Under certain atmospheric conditions the rate of chemical reactions increases, a rapid growth of impurities concentrations takes place and photochemical smog forms. As laboratory and field researches have shown, one of the products of photochemical smog is ozone. Its concentration grows parallel with the growth of secondary aerosols and number of gas impurities. It can reach great values. A sharp rise of  $O_3$  in daytime hours was often observed in cities.

The purpose of this paper is to elucidate conditions under which intensive formation of photochemical smog takes place on the basis of systematic diurnal  $O_3$  measurements in the surface layer of the atmosphere in Tbilisi. A statistical analysis of  $O_3$  dependency on the atmosphere's characteristics at various hours during day and night, including autocorrelation analysis of parallel time series of  $O_3$  and the atmosphere parameters were carried out. Conclusions are made of atmospheric processes influencing upon quantitative contents of this pollutant in night time and day-time with different duration of solar radiation. Analysis is made of conditions accompanying the cases with typical for photochemical processes diurnal dynamics of  $O_3$ . In the majority of these cases such atmospheric conditions took place that a sharp growth of  $O_3$  in afternoon hours could not be caused by horizontal advection or vertical transfer from upper layers of the atmosphere. So during the days under consideration intensive photochemical processes took place which led to local formation of ozone and other products of chemical reactions. On the basis of investigations and the results of other authors works conclusions are drawn about conditions favouring the formation of secondary pollutants of the atmosphere, the height of their generation and the time of maximum intensity of photochemical processes in the lower troposphere.

TERPENES, OZONE AND NITROGEN OXIDES  
IN A FINNISH BACKGROUND STATION

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Photochemical formation of ozone requires presence of  $\text{NO}_x$  and hydrocarbons. Natural emissions of hydrocarbons as terpenes are large, which directs considerable interest in studying their role in ozone procedss.

As previous terpene measurements point to large variation in their concentrations, existing data on background concentrations of these compounds cannot be used for exact analyses. Multicomponent data are required for the purpose.

The terpenes, ozone and  $\text{NO}_2$  in a coniferous forest were determined in spring, summer and fall in the same stand locations. At the same time meteorological and some aerosol data were registered. The largest temperature difference between the periods was 40 °C. The terpenes from a Tenax-sorbent sampling were analyzed using a thermal desorption and mass selective gas chromatography.

The terpenes and ozone showed similar seasonal variation with high values in July. Their diurnal variation was inverted, with terpenes highest at nights and ozone lowest.

Ozone was effectively reduced inside the forest (at 10 cm from the closest tree stem) at nights when the temperature is high, but in principal not at all when the temperature is below zero centigrades.

The ratio terpenes/ $\text{NO}_x$  was high only during nights. Such values for the ratio  $\text{CH}/\text{NO}_x$  ( 5-10) have in laboratory tests produced maximal ozone concentrations. The terpene-ozone- $\text{NO}_x$  phenomenon is more properly analysed in the work.

CASE STUDIES OF THE CHEMICAL COMPOSITION OF ATMOSPHERIC AEROSOL  
AT THE SZRENICA SUMMIT, KARKONOSZE RANGE, POLAND

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The concentrations of TSP,  $\text{SO}_2$ ,  $\text{NO}_x$ , sulphate aerosol, trace metals and PAN in atmospheric aerosol samples have been measured since April 1986 at the Szrenica summit 1362 m a.s.l. for air masses coming from south- and north-western Europe. Samples were taken in 24 hour intervals continuously for seven days every month. The sampler for sulphate and ammonium aerosol consisted of a prefilter and a bubbler with hydrogen peroxide and sulphuric acid solution, respectively. Microscopic analysis (TEM, SEM) was used to study the physical and chemical properties of aerosol microparticles.

EM examinations have revealed large quantities of extremely fine particles (radius below 0.1  $\mu\text{m}$ ). Concentrations of fine sulphate aerosol determined from the hydrogen peroxide solution after subtracting sulphur concentrations associated with  $\text{SO}_2$  were also found to be very fine, amounting up to 200  $\mu\text{g}/\text{m}^3$ . Conversion of  $\text{SO}_2$  to  $\text{SO}_4^{2-}$  ranged from 8 to 76%, according to the history of air masses, cloud activity and the height of the inversion layer. A reasonable explanation of these results is that a substantial portion of  $\text{SO}_4^{2-}$  has formed in the region of interest and does not come from long-range transport.

Based on aerosol chemistry and meteorological data, source apportionment was carried out for the remaining investigated species.

## SIMULATION OF AEROSOL PARTICLES IN A LARGE SMOG CHAMBER

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The principal problem describing the behaviour of atmospheric aerosols consists of the simultaneous action of both physical and chemical processes. The physical processes comprise linear processes like the particle deposition and non-linear processes like coagulation. The simulation of such a system is unfortunately not possible from a strictly theoretical basis but need quite a few parameters which have to be determined experimentally. The characteristic time is quite different for the different processes and experimental methods are requested suitable to the specific problem. Fast processes are preferably investigated by flow reactors, but slow processes like a number of heterogeneous reactions on particle surfaces have to be examined in large batch reactors since the residence time of the particles is large enough only in such reactors. To plan such an experiment a number of calculations with the computer code NACHE were performed which is able to simulate the behaviour of an aerosol system under the influence of all important aerosol physical processes (coagulation, nucleation, condensation and deposition processes) and a limited number of chemical processes like formation of gaseous sulfuric acid and a special type of heterogeneous oxidation of sulfuric dioxide on the particles. It can be shown which processes of which characteristic time and reaction velocity can be investigated by the planned experiments in the 80 m<sup>3</sup> container. The findings which processes can be examined does not only depend on the particle size but also on the thermodynamic conditions like temperature and pressure of the carrier gas which might be important if stratospheric aerosols have to be investigated.

A NOVEL INSTRUMENT FOR THE CLASSIFICATION OF AIRBORNE PARTICLES  
ON THE BASIS OF SIZE, SHAPE AND COUNT FREQUENCY

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A laser light scattering instrument has been developed to assist in the study of airborne particulate material. The instrument employs a novel optical scattering chamber design which allows airborne particles to be drawn in from an ambient atmosphere and examined individually at high throughput rates (10,000 per second). The combination of a system of optical detectors and high speed data processing allows in the classification of the particles in terms of their size (or for non-spherical particles, the spherical equivalent size), their shape (based upon the spatial intensity distribution of the scattered light), and their time-of-flight through the laser beam (which may later be related to aerodynamic size). The instrument is designed for particle detection in the size range 1 to 10  $\mu\text{m}$ , although this range may be altered if necessary.

Raw data from the individual particles is stored in real-time for post sampling analysis and display. During the analysis, particles are ascribed an Asymmetry Factor, or asphericity factor, which relates to the degree of asymmetry in their spatial light scattering profiles. Thus spherical particles have factor 0 and fibrous particles factor 100. Cumulative particle data plots may then be displayed showing particle Size vs. Frequency, Asymmetry Factor vs Frequency, and perhaps most interestingly, Size vs. Asymmetry vs. Frequency. The three-dimensional surface produced by the latter process represents a characteristic signature of the ambient aerosol, and early results indicate that possibly pertinent features, such as, for example, the presence of a low concentration of spherical particles (or droplets) in a non-spherical particle population, or the presence of fibrous particles, may be determined from the surface topology.

Current work is examining the degree of orientation experienced by non-spherical particles in the laminar-flow system used for particle delivery into the chamber, and on the scattering properties of a variety of particulates including salt crystals, coal dusts, industrial fibre dusts, and some naturally occurring pollens.

### SAMPLING ARTIFACTS FROM CURVED AIRBORNE INTAKE SYSTEMS

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Although many investigators have collected aerosols from airborne platforms, the passing efficiency of the common curved- tube intake systems has not been carefully measured under actual flight conditions. We will report on in-flight comparisons between a variety of intake geometries, including one which has no curved intake tube. Our preliminary data suggest that the losses in curved intakes are far larger than we had previously imagined, so that the data from earlier programs may in fact be lower limits to actual aerosol concentrations.

A STUDY OF PARTICULATE RESUSPENSION FROM AN ASH-DUMP

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Measurements were carried out of the mass concentration of particles resuspended from an ash-dump of coal-fired power plant as well as a wind velocity at the height 1.5 m.

The threshold wind velocity  $v_t$  was obtained as about  $2.3 \text{ ms}^{-1}$  from the relation of aerosol concentration as a function of windspeed. These investigations were performed by use of the Model 3500 Respirable Aerosol Mass Monitor - Piezobalance at 200 m downwind distance from the dump.

The simple box model was also studied in detail by applying it to a dust resuspension from the ash-dump of coal-fired power plant. From these considerations the surface emission values as a function of windspeed were estimated. Based on the measurements of aerosol mass concentrations made with the Staplex High-Volume Samplers, calculations predict surface emissions as  $0.98$ ;  $1.35$  and  $28.01 \text{ mgm}^{-2}\text{s}^{-1}$  for the windspeed  $5, 6$  and  $10 \text{ ms}^{-1}$  respectively.

These results indicate that the model can provide information necessary for the study of the influence of individual surface resuspension sources on the lower atmosphere.

APPLICATION OF AN ISOTHERMAL HAZE CHAMBER TO STUDIES OF MIXED  
SOLUBLE/INSOLUBLE ANTHROPOGENIC PARTICLES

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Anthropogenic aerosol particles such as those resulting from petroleum fuel combustion have been found to be composed of elemental carbon, organic carbon, and sometimes internally mixed water-soluble compounds. Particles that are mostly water soluble, and are of sufficiently large size, are likely to be activated as atmospheric cloud condensation nuclei (CCN), and are subject to relatively rapid atmospheric removal. Smaller particles, or those containing only marginal amounts of water soluble compounds, will be resistant to activation and "nucleation scavenging", and thus are candidates for longer residence times in the atmosphere. These anthropogenic nuclei may be detected at remote sites, in low concentrations, where water nucleation measurement techniques provide one method of distinguishing them from other mixed or mostly soluble particles. This differentiation is clearly demonstrated on a size-critical supersaturation (Sc) plot, generated by electrostatic size classification (EC) of the sample particles, and subsequent measurement of their Sc spectra by means of an isothermal haze chamber (IHC).

In this application, IHC devices have an advantage over other CCN detectors in that the difference in behaviour between sparingly soluble and mostly soluble particles is accentuated by the application of the 1.00 (or less) saturation ratio in the IHC, as opposed to the supersaturations applied in other types of CCN detectors. For example, the equilibrium diameters of the particles in the two types of chambers may differ by as much as a factor of ten. A measurement system consisting of an EC/IHC combination has been built and carefully calibrated using pure soluble monodisperse particles. The chamber operates at RH lower than 100% by using saturated solutions. Data obtained with this system, when applied to typical fuel smoke particles, will be presented.

MICROPHYSICAL CHARACTERISTICS EVOLUTION OF THE NATURAL AEROSOL  
JUST BEFORE FOG FORMATION

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Air samplings realized in conditions where fog formation was possible revealed the microphysical reality of a particularly coherent continuity between:

- 1) the cloud condensation nuclei (visual range conditions well above 1 km), the detection and study of which could only be achieved by an isothermal haze chamber with respect to their condensation activity (supersaturation spectra), and
- 2) the nuclei droplet (haze conditions with visual range around 1 km) and/or the fog droplets induced by the preceding nuclei (visual range below 0.8 km), all particles detectable with the help of a size spectrometer.

In other respects, the data of the haze chamber used as above in 1) have been quite representative of a further fog formation in as much as the relative humidity of the air reached at least 100%.

PERIODICITY OF EXTINCTION CURVE OF DIELECTRIC SPHERICAL PARTICLES

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We have shown that in the large size parameter region ( $x \gg n^2/2$ ) the  $Re(a_n + b_n)$  is independent of the summation index  $n$ . For real refractive indices  $m < 2.5$  the dominating term of  $Re(a_n + b_n)$  has the  $x$ -dependence of the form  $\sin^2[(m-1)x]$  leading to the periodicity of  $Re(a_n + b_n)$  and of the extinction curve  $Q_{ext}(x)$  given by  $\Delta x = \pi / (m-1)$ . The derived periodicity is the same as the periodicity of interference structure derived using the anomalous diffraction condition  $m-1 \ll 1$ . At refractive indices  $m > 2.5$  the extinction curve does not have a simple periodic structure, since several terms of approximately equal magnitude and different  $x$ -dependence contribute to the  $Re(a_n + b_n)$ .

OBSERVATIONS OF A POSSIBLE GOBI DUST EVENT BY INFRARED  
LIDAR NEAR MAUNA LOA, HAWAII

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During November and December 1988 NOAA's Doppler Lidar was stationed near the GMCC climatic observatory on the slope of Hawaii's Mauna Loa volcano. We were participating in a NASA sponsored experiment to measure vertical aerosol backscattering profiles at 10.59 and 9.25 microns wavelength under clean, or background, conditions, and to compare our measurements with in-situ samplers. In this regard the experiment was very successful. But on December 2 we also observed highly enhanced aerosol loading in several elevated layers for tens of hours. These layers advected over us from the east, then settled through the altitude of the lidar and the observatory (approximately 3.5 km ASL). In this paper we will analyze the temporal and spatial development of the event as observed by the lidar, and include data that were acquired simultaneously with several types of particle samplers from GMCC, Georgia Institute of Technology, and the University of Hawaii. We hope also to include back-trajectories of the air parcels in question for indication of the aerosols' source.

AEROSOL BACKSCATTERING CLIMATOLOGIES AS OBSERVED BY  
INFRARED LIDAR

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We present analyses of several years of lidar backscatter data acquired near Boulder, Colorado at 10.59 microns wavelength, and of one month of data acquired in Fall 1988 in clean Pacific air near Mauna Loa, Hawaii at 10.59 and 9.25 microns wavelength. The continental profiles include data before, during and after the El Chichon event, from which we deduce purging time constants on a global scale for both the stratosphere and the troposphere, and hypothesize that tropopause folding is responsible for significant amounts of stratosphere/troposphere exchange. In contrast, the more recent continental and Pacific Statistical analyses of the data show that many of the backscatter values are log-normally distributed, indicating single sources and proportional diffusion of aerosol-laden upper air parcels.

AN INTERACTIVE MODEL FOR PREDICTING SCATTERING FUNCTIONS  
OF POLYDISPERSE AEROSOLS

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Details of a PC-based computer model for predicting optical properties of polydisperse aerosols are presented. This program permits the user to compute extinction and angular scattering functions for aerosols of arbitrary size distribution and illuminating light spectral properties. The effect of relative humidity on optical properties can be included. Calculations are carried out in an efficient manner with a minimum of delay between input and output. The program is available for general distribution.

CONCENTRATION OF SOME AEROSOL COMPONENTS AND ITS INFLUENCE  
ON GLOBAL SOLAR RADIATION ABSORPTION AND SCATTERING  
IN CENTRAL PART OF UPPER SILASIAN INDUSTRY REGION, POLAND

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The work will be based on simultaneous measurements of aerosol components like elemental carbon, sulfates and nitrates and actinometric parameters, direct and total global solar radiation, which have been carried out during approximately 2.5 years, 1987 - 1989 on Katowice - Zaleze station.

In the paper will be preliminary presented reciprocal relationships between mentioned aerosol components and its influence on absorbed and scattered shortwave solar energy reaching the earth's surface.

BACKGROUND AIR QUALITY AND DEPOSITION OF TRACE ELEMENTS  
IN THE PROVINCE OF SOUTH-HOLLAND

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Recently a survey was made of the aerosol/trace element situation in South-Holland, the Netherlands, consisting of an emission inventory and measurements of the background air quality and background deposition values, following a general survey of the background air pollution in the whole country. Use was made of high volume samplers, medium volume samplers, cascade impactors and moss.

There are no large man-made sources of trace elements in the province. Relatively large emissions come from oil refining, waste combustion and traffic. In fact, the emissions in the province, as well as in the whole country, are very low compared to the emissions in the surrounding countries. The absence of large sources is reflected in the values of the (background) air pollution. Analysis of the measured values shows that the greater part of the aerosols comes from the surrounding countries.

For cadmium a further analysis was made: the total economic and environmental input, accumulation and output were analysed. It was found that the deposition of cadmium alone exceeds the decrease of cadmium in the soil from harvesting crops and leaching. Even with an estimate for the fraction of cadmium from natural sources, such as soil dust and sea spray, the deposition of cadmium from man-made sources still only just about balances the decrease of cadmium in the soil. This means that where no crops are harvested, as in nature reserves, or where fertilizer, manure, sewage sludge or compost is used, a steady increase of cadmium in the ground, and hence in vegetation and organisms is foreseen. As it is predicted that cadmium accumulates in the economic cycle and more waste incineration will take place, the (background) air pollution of cadmium, and probably of several more trace elements, will increase, unless preventive measures are taken. The continuing increase of cadmium in the environment will, in due time, become a threat to the ecological systems and to the economy.

STUDY OF ATMOSPHERIC AEROSOLS IN A TERRAIN-INDUCED NOCTURAL  
BOUNDARY LAYER USING BISTATIC LIDAR

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The aerosol content of the lower atmosphere fluctuates under the continuous influence of particle source and deposition mechanisms. These fluctuations in aerosol amount, particularly in the atmospheric boundary layer can easily be monitored by means of lidar as the scattering from aerosol particles contributes a maximum to the lidar backscattered intensity in this region. Thus the inhomogeneities in ambient aerosol content can be used as tracers of the structure and stratification of the nocturnal boundary layer which can be attributed to the phenomena of convective activity and turbulence. Better height resolution can be achieved with lidars and these instruments have been in use for investigating small-scale atmospheric phenomena.

A bistatic lidar has been developed at the Indian Institute of Tropical Meteorology, Pune, India for sensing lower atmospheric aerosol characteristics remotely. The measuring site is surrounded by hillocks which affect the aerosol transport particularly in the lower levels. The aerosol vertical profiles (20-1000 metres) obtained during night-time between 2000-2130 hrs IST to 20 June, 13 October, 11 November and 8 December 1988 have been presented in this paper. These profiles have been measured at a height interval of 20 metres to study the stratified aerosol layered structure. Moreover, normalised concentration gradients (NCG) have been computed from these profiles to extract information on layer characteristics. It is observed that the gradient was very steep up to 150 metres on all the four experimental nights. The profile measured in June showed thin layers throughout the height region while those obtained in the other three months exhibited relatively smooth altitudinal variation. However, the overall concentration of aerosol particles in the lowest part of the atmosphere was found to be high in October, November and December when compared to June which corroborates the earlier studies of aerosols at this station. In order to examine the altitude structures, the above aerosol profiles are compared with the profiles of radiometersonde derived temperature and balloonsonde derived winds. The results are explained on the basis of terrain effects and stratified turbulence that prevailed at the station on the above four days.

VERTICAL PROFILE AND ELEMENTAL CONCENTRATIONS  
OF AEROSOLS IN A FOREST AREA

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During summers 1987 and 1988 four one week period stacked filter unit measurements were carried out in Hyttiala, area which can be considered as background area in Finland. The sampling periods varied from 12 h to 2 d. The size distributions of aerosols were determined during sampling periods with ten stage HAUKE low pressure impactor. The mass concentrations of airborne particles were measured both in fine (aerodynamic diameter  $< 1 \mu\text{m}$ ) and coarse ( $> 1 \mu\text{m}$ ) fractions using Nuclepore filters. Stacked filter units were situated in a meteorological tower at heights 3, 6 and 9 m which made it possible to obtain the vertical profile of mass and element concentrations. The top of the tower was 3-4 meters above the forest canopy. Also meteorological data were registered simultaneously. The particulate matter was analysed using the elemental analysis method PIXE.

Mass distributions show that particle matter is divided between the fine and coarse fraction but elemental analysis show that certain elements like K, Ca, Si and Fe appear quite differently in these fractions. The most intense element analysed by PIXE was silicon.

Vertical profiles for the elements differ also. Si and Fe have sources in the ground as their concentrations systematically decrease when going from ground to top. S in fine particle fraction varies much with time reflecting transport from outside but it also may have sources inside the forest. Strikingly, S was high during nights. K and Ca have no systematic profile so they have sources inside the forest. These sources are varying, most probably with the meteorology. It was remarkable that Cl was found inside a forest in the site located far from coast. As a conclusion internal cycling of elements inside the forest has an important role when analysing background pollution.

MODELLING DRY DEPOSITION OF PARTICLES TO THE OCEAN

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It is well known that some of the particles in the ocean mixed layer derives from the atmosphere, but it is generally unknown how large the atmospheric input is. A resistance model to describe the transport of particles to the ocean has been developed. The transport is expressed in terms of the deposition velocity,  $v_d$ . No chemical or biological processes is included in the model, but only physical processes. The processes, which is included in the model is sedimentation, including hygroscopic growth of wet particles, Brownian diffusion, atmospheric (turbulent) diffusion and wash out by spray-droplets from whitecaps. The model operates in three different layers: a turbulent well mixed layer, a laminar sublayer just above the smooth surface and a turbulent sublayer just above the whitecaps. The purpose of making this model is to provide a tool for a critical evaluation of the relative importance of the individual processes. At this stage, it is therefore a model which is primarily intended to serve as a basis for theoretical discussions.

Of the included parameters the wind speed,  $U(z)$ , primarily through its effect on the area with whitecaps, the particle diameter, and the particle density, is found to be the most important.

MODELLING OF THE TRANSPORT OF WATER-SOLUBLE AEROSOLS BY A  
TROPICAL SQUALL LINE

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A two-dimensional eulerian cloud-model is used to study the evolution of a tropical squall line and the transport of water soluble aerosols. The basis of the meteorological data are observations during the COPT-experiment in Africa. The content of the cloud water, rain water, cloud ice and the wind components are computed over a period of 120 minutes in a domain of 40 x 15 km with a vertical and horizontal resolution of 333 meters. Substances, which are emitted at the ground, are transported up to 12 km by the strong updraft motion and are distributed in the horizontal direction. It will be shown, that there are high concentrations of ground emitted substances in the cloud level. The important role of correct parameterization of cloud microphysics on the evolution of the cloud and on the modelling of the scavenging and transport of water soluble aerosols are discussed.

TRANSPORT, CHEMISTRY AND DEPOSITION OF AIRBORNE GASEOUS  
AND PARTICULATE POLLUTANTS IN A MARINE ENVIRONMENT

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We report on shipborne measurements carried out during a cruise across the Baltic Sea in April-June 1985. Concentration levels of both gaseous ( $\text{SO}_2$ ,  $\text{HNO}_3$ ,  $\text{NH}_3$ ,  $\text{Hg}$ , PAH) and particulate ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , PAH) compounds indicate that the Baltic Sea cannot be considered as a remote clean area. Meteorological patterns explained much of the time evolution of the concentrations. The ratio  $\text{SO}_2/\text{SO}_4^{2-}$ , related to the conversion rate of sulphur dioxide to particulate sulphate, was generally smaller than one and depended on relative humidity and ammonia concentration. The high correlation between sulphate, nitrate and ammonium ions can be explained by their anthropogenic origin and their occurrence under the form of salt. The probability for ammonium nitrate particles to exist in the meteorological conditions prevalent during the cruise, was found to be weak. The experimentally determined ammonium nitrate dissociation constants were in fair agreement with thermodynamical predictions, with the exception of clean air cases. Ammonia was found to have an uncertain path with possible marine sources, a weak correlation with other compounds, and a variable residence time. These data, completed with corresponding observations from coastal EMEP-stations, allowed us to assess the dry deposition rate and the chemical transformation rate coefficients in a simple transport-scavenging model for sulphur compounds. The results were satisfactory for the dry deposition velocity, and agreed with the predictions of a theoretical model, but were conflicting for the transformation rate, and very sensitive to the various assumptions made in the computations.

IONIC BALANCES IN MARINE RAINWATER WITH A SPECIAL EMPHASIS  
ON SOURCES OF ALKALINE AND ACIDIC SPECIES

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Rainwaters were collected in ultra clean conditions during three field experiments (two in Corsica Island and one during a North-South Atlantic transect of the 1988 "Polarstern" cruise). These rainwaters are immediately filtered and the major ions were measured by ionic chromatography, atomic absorption and colorimetric methods.

In these marine areas, the concentrations of major ions range over one or two orders of magnitude and are generally dominated by the redissolution in rainwater of the sea salts. The non-sea salt component in rainwater shows the presence of the anthropogenic nitric and sulfuric acids which are well correlated with the origin of the air masses and which lead to acidic rains (pH from 4 to 5).

High values of pH (6-7) are also observed and can be explained by the neutralization of strong acids by natural alkaline dust as calcite, which are mainly transported from the desert areas of the African continent. A study of these ionic balances in relation with 3D air mass trajectories allows one to demonstrate the composition of these two components during African dust transports.

CONCENTRATION PROFILES OF TRACE ELEMENTS OVER THE NORTH SEA  
AND THE ATLANTIC OCEAN: ANTHROPOGENIC AND NATURAL CONTRIBUTION

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During a 1988 "Polarstern" cruise, eighteen aerosol samples were collected over the North Sea and the Atlantic Ocean (between 53°N and 32°S) in ultra clean conditions. Measurements of trace elements (Al, Si, P, S, Mn, Zn ...) were performed by X-Ray fluorescence.

The latitudinal profile exhibits a strong decrease of particulate concentrations for pollutants as Zn or S\*. In the North Sea, concentrations as high as  $3500 \text{ ng.m}^{-3}$  of sulfur and  $50 \text{ ng.m}^{-3}$  of zinc were observed while the concentrations are in the order of  $500 \text{ ng.m}^{-3}$  and  $1 \text{ ng.m}^{-3}$  respectively in the Southern Hemisphere and Tropical North Atlantic. These results point out the strong influence of the anthropogenic emissions of the North European countries on the atmosphere of the marine areas.

On the contrary, soil derived particles (as indicated by Al or Si concentrations) show high concentrations between 40°N and 10°N. 3D air mass trajectories indicate that such high concentrations over the Tropical North Atlantic are related to Saharan dust events.

Obviously, elements associated to both anthropogenic and crustal sources (P and Mn) presented a mixed profile of concentrations.

DIURNAL VARIATION OF THE CONCENTRATIONS OF RADON AND ITS  
SHORTLIVED DAUGHTERS IN THE ATMOSPHERE NEAR THE GROUND

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In connection with deposition studies of aerosol on vegetation, besides the size distribution of the aerosol the vertical turbulent diffusivity is the most important meteorological parameter, which influences the behaviour of the aerosol particle in the surface layer of the atmosphere. From the vertical profile of the radon concentration and the state of equilibrium between radon and its daughters above the canopy the vertical turbulent diffusivity can be calculated.

The diurnal variation of radon (Rn-222) and the radon daughters (Po-218 (RaA), Pb-214 (RaB, Bi-214 (RaC) concentrations have been measured simultaneously at 0.2, 0.7, 1.2 and 3.2 m above the ground. At the same positions the meteorological parameters, temperature and wind velocity, were continuously registered. The variation of the concentration profiles and the derived values for the turbulent mixing during a day and at different weather conditions are discussed.

REMOVAL OF AIR BORNE PARTICLES BY RAIN DROPS

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A mathematical model was developed for removal of airborne particles by rain drops. In this model separation of small particles by inertia and diffusional mechanisms are considered on individual drops. Then a material balance over a volume element was carried out in order to take into account the effect of rainfall and wind velocity. The results indicate that the removal rate increases for particle above 0.2 micrometer as the particle size increases. For particles below 0.2 micrometer the trend is reversed and the removal rate increases as the particle size decreases.

CONFERENCE PAPER PRESENTATIONS

(CONTINUED)

## LONG RANGE TRANSPORT OF AEROSOLS

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Across Europe, rather usual acidic precipitation is reported. The average pH-value is slightly more than 4 with a considerable standard variation. As usual, such an average is only half the story. A close inspection of the data set available in Mainz shows a two peaked frequency distribution with an acidic peak as mentioned above and a peak around pH=6, well on the basic side for rain. Basic rain is reported in pronounced cases of airmass transport from the South-West and furthermore from Northern Africa. Obviously, this is Saharan air. With multiday backward trajectory analysis for all the other cases of basic pH, the trajectories always pass over the Saharan desert. For Central Europe, the latest case occurred on May 1, 1989. Saharan air arrived from the North-East having travelled more than 5 days over Eastern Europe and a distance of more than 4500 km. Transport over such a long distance is a regular phenomenon for Saharan dust across the Atlantic ocean towards the Caribbean Sea.

Mineral dust is an excellent tracer for long range transport, because it is hardly modified during transport. Radioactivity attached to aerosol particles is of equal use, and the Chernobyl accident case has shown a transport distance equal to that of the Saharan desert dust.

There are many isolated reports of long range transport of specific aerosol substances in the past. Under stratospheric conditions in the absence of clouds and precipitation, such a long transport seems to be easy, as the volcanic eruptions with the worldwide spread of dust and the drastic effects on temperature and light scatter show.

For aerosols, the troposphere usually seems to inhibit such a long transport. Usually for the troposphere spread and dilution of material is assumed to lower quickly and drastically the concentration of freshly released trace substances thus reducing its effects. So only very substantial explosions of material could be detected far downwind. There are indications that the tropospheric spread and dilution is not so omnipotent as usually assumed, thus keeping an aerosol cloud together and delivering rather high concentrations of aerosols to far away places. As an example, the water vapour of the atmosphere with a residence time of 10 days is comparable to that of the natural aerosol. The confinement of water clouds by thermodynamical atmospheric processes can be seen visually and on satellite images in the infrared. A long distance transport of aerosol clouds certainly has an influence on our understanding of background aerosols and their existence in an atmosphere faced with an increasing number of anthropogenic aerosol sources and substances. This question has never been investigated systematically, but certainly deserves more attention for promoting our understanding of the natural aerosol and the atmosphere.

TROPOSPHERIC AEROSOL SIZE DISTRIBUTIONS IN EASTERN AND NORTHERN  
NORTH AMERICA BETWEEN 1982 AND 1988:  
A SIGNATURE FOR TROPOSPHERE-STRATOSPHERE EXCHANGE?

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The size distribution of the 1000-500 mb tropospheric aerosol was characterized at 5 North American locations from 1982 until 1988 using light scattering instruments. The instruments, a Particle Measuring Systems ASASP-100X and a PMS FSSP-100, were mounted under one wing of a Twin Otter aircraft. This ASASP measures particles between about 0.17  $\mu\text{m}$  and 2.0  $\mu\text{m}$  diameter, and this FSSP measures particles between about 1  $\mu\text{m}$  and 35  $\mu\text{m}$ . The data set is comprised from 7 studies each of 4-9 weeks duration: central Ontario, June-July 1982; central Ontario, January-February, 1984; upper New York state, October-November 1984; Nova Scotia, January-March 1986; the Canadian high arctic, April 1986; southern Quebec, February 1987; and central Ontario, July-August 1988. Except near primary source regions (e.g. oceans) particles in the range 0.17-0.5  $\mu\text{m}$  generally dominate the size distributions, to the extent that the distributions are more nearly monodisperse than previously indicated (the range of the geometric standard deviation of the particles between 0.17  $\mu\text{m}$  and 0.7  $\mu\text{m}$  ranges is 1.22-1.26). When flying through clouds reductions in the number concentrations of these particles correlate very strongly with increases in cloud droplet number concentrations, indicating these particles are responsible for shaping the microphysical character of liquid-water clouds. A secondary mode near 0.7  $\mu\text{m}$  is present in many of the later winter and spring spectra when the total number concentration of particles is very low. The evidence is constant with this being a real phenomenon rather than an artifact of light scattering. Some of the possible explanations for the mode near 0.75 include marine aerosols, desert aerosols, or simply the evolution of the particle spectra via wet scavenging, coagulation, settling, and gas-to-particle conversion. Most of the evidence, however, points to the stratosphere as the source. If these particles can be verified as originating in the stratosphere, then this may provide a new tool for studying the exchange of air between the troposphere and the stratosphere.

AEROSOL MEASUREMENTS IN THE SOUTHWEST PACIFIC

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Measurements of the physical characteristics and chemical composition of the atmospheric aerosol have been made around New Zealand and other southwest Pacific Islands from an instrumented aircraft between 1985 and 1988. Latitudes ranged from 46 °S to 1.5 °N at altitudes of up to 6 km. Single particle optical counters were used to cover the size range 0.1  $\mu\text{m}$  to 16  $\mu\text{m}$  diameter.

The results showed that in the free troposphere aerosol concentrations ranged from 2  $\text{ml}^{-1}$  to 50  $\text{ml}^{-1}$  and the size distributions could be characterised by a Junge slope of around - 3.5. Most of the aerosol was dry or hydrated sulphur containing particles. Potassium containing particles were also found over a wide range of latitudes.

In the planetary boundary layer the concentrations were generally greater and more variable and the size distributions often bimodal.

VERTICAL PROFILES OF AEROSOL PROPERTIES  
IN THE ARCTIC SUMMER TROPOSPHERE

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The Arctic summer troposphere probably is the cleanest region in the northern hemisphere. In cloud-free surface air, total particle concentrations down to  $1/\text{cm}^3$  and total mass concentrations down to about  $100 \text{ ng m}^{-3}$  have been measured and the major chemical components of this aerosol have been determined. Very little, though, is known about the vertical distribution of aerosol properties in this remote region.

In connection with the Marginal Ice Zone Experiment (MIZEX-84) a large number of flight measurements were made in the Framstrait area during June and July, 1984. With an aerosol payload onboard the German research aircraft Falcon, total number concentrations and integrating nephelometer measurements were performed from the boundary layer up to six kilometer altitude. Simultaneous temperature, humidity and cloudphysical measurements were used to exclude cloud passages and high humidity situations from the data. From the cleaned data set average vertical profiles and confidence intervals of number concentration and particulate light scattering were calculated.

The results show rather even concentrations from the ground up to six kilometers. Layers with elevated concentrations were found at about 1.5 km (level of most frequent long-range transport), four km (local inversion) and six km (near the tropopause inversion). With three-dimensional back trajectories from the flight area, pathways and origin of the air masses will be explored in our presentation.

VERTICAL PROFILES OF AEROSOL TO 7 KM IN THE ARID SOUTHWESTERN US

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Concentrations of natural occurring aerosol were measured to 7 km altitude using aircraft-borne light scattering aerosol counters during June and September of 1988. Particles with equivalent diameter 0.2-10 micrometer show little vertical variation in the mixing layer (below about 4 km); above the mixed layer a significant decrease in concentration is observed throughout the size range. Decreases in concentration above the mixed layer are typically a factor of 5 to 10 for June, and for September aerosol concentrations decrease two orders of magnitude from the top of the mixed layer to 7 km. Aerosol concentration vertical profiles are compared to previous measurements of Fitch and Cress (1), Kondratyev (2), Krekov and Rakhimov (3), Rosen et al (4,5) and Rosen and Hofmann (6).

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ATMOSPHERIC TRANSPORT OF SULFUR AND NITROGEN ACROSS THE  
ATLANTIC OCEAN FROM NORTH AMERICA TO EUROPE

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This paper addresses the subject of the long range transport of sulfur and nitrogen from North America to Europe.

The results of the Western North Atlantic Ocean Experiment [WATOX] are used to show that about thirty percent of the S and N emitted to the North American atmosphere is transported eastward to the atmosphere of the western North Atlantic Ocean. Of this material, up to fifty percent is transported to the North Atlantic Atmosphere east of 60W.

The impact of these emissions on the composition of the central and eastern North Atlantic Ocean atmosphere is examined using eight years of data on the composition of precipitation from Bermuda and four years of data from the west coast of Ireland at Adrigole.

In conjunction with this analysis of precipitation chemistry data, we also use information on the emissions of S and N to the European atmosphere together with estimates of the eastward transport of these species from Europe to the eastern North Atlantic atmosphere.

Significant findings are that the composition of precipitation over most of the North Atlantic Ocean is contaminated with anthropogenic emissions. Over the western North Atlantic Ocean, the sources are predominantly North American. Over the eastern North Atlantic Ocean, the sources are predominantly European.

LONG-RANGE TRANSPORT OF TRACE METALS TO  
NORTH EUROPEAN MARGINAL SEAS

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Within the framework of a research contract with the German Federal Environmental Agency the long range transport of trace metals over Europe and the deposition into the North Sea and the Baltic Sea are estimated using the EMEP-(European Monitoring and Evaluation Programme) trajectory model. The results for the total atmospheric deposition into the North Sea and the Baltic Sea show, that the calculated values are lower than previous estimates based on extrapolations from measurements at coastal sites and ship measurements in the southern Baltic Sea, respectively.

To verify the model results calculated atmospheric concentrations of lead were compared with observed concentrations at two locations at the German coast of the North Sea and Baltic Sea. Good agreement between the measured and calculated concentrations was obtained for these two locations relatively close to the main emission areas in Central Europe suggesting that the emissions are accurately assessed and that the model is able to predict the total deposition into the entire sea areas reasonably well.

Currently the model verification is extended by a comparison with measurement data from 9 locations along a meridional cut ranging from Birkenes in southern Norway to Ny Alesund, Spitsbergen, in the Norwegian Arctic. Preliminary results indicate that the model tends to underpredict atmospheric concentrations remote from the major emission areas. It is expected that the planned refinements in the parameterization of the deposition processes will improve the results.

Moreover, an attempt is made to identify natural background concentration levels at measurement stations in the Norwegian arctic. This is done by using calculated 96 h-backward trajectories for a classification of air samples with respect to emission areas. During episodes when trajectories arrive from areas where no emissions occur the corresponding measurement values are considered to be the natural background level and to be used as input data for the model.

SOURCE, TRANSPORT AND DEPOSITION OF LEAD IN THE  
MEDITERRANEAN ATMOSPHERE

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Beginning in 1985, a continuous aerosol sampling program has been undertaken at a coastal location in Northwestern Corsica (Western Mediterranean sea). Twenty-four hours aerosol samples were collected on 0.4  $\mu\text{m}$  pore size filters from a 10 m high tower. Total deposition (wet + dry) samples were collected during the same period with a time step of 15 days.

We only present in this paper the results concerning Pb. The temporal variability of atmospheric particulate lead concentrations is discussed on various time scales (daily, seasonal, annual). The source-regions and the atmospheric pathway of particulate lead are identified by using 3D air-mass trajectories.

These factors, which are able to explain abrupt changes in atmospheric concentrations of lead, are also used, in addition to precipitation rates, to understand the variability of atmospheric deposition of lead over the Northwestern Mediterranean.

EUROPEAN SOURCE AREA IDENTIFICATION AND APPORTIONMENT  
OF LONG RANGE TRANSPORTED AEROSOL

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In order to assess the impact of various European source areas on the ambient aerosol in southern Sweden, samples were collected during the period from April 1985 to December 1986 at three background stations situated on small islands along the south-eastern coast of Sweden. A specially designed, two-size fractionating, low-volume, microcomputer-controlled sampler suitable for automated aerosol sampling in remote locations was utilized. It allows for four weeks of continuous operation, with 12-hour sampling periods. Some 600 of the fine fraction samples ( $D < 2 \mu\text{m}$ ) were selected for analysis, based on the 96-hour air mass back trajectories. The elements S, K, Ca, Ti, V, Mn, Fe, Ni, Cu, Zn, Br and Pb were detected in the PIXE-analysis, and Na was analysed simultaneously taking advantage of the proton-induced gamma-ray emission. Prior to these analyses, a light attenuation technique was used for the analysis of elemental carbon (soot).

The wealth of data produced was subjected to a multivariate statistical analysis, mainly using the classification method SIMCA (Soft Independent Modelling of Class Analogy) and the regression method PLS (Partial Least Squares regression). Applying these methods on the data set showed that the long-range transported aerosol originating from various larger European source areas have different chemical composition, and the observed differences are statistically significant. This enables us to perform a source area-receptor modelling on the long range transported aerosol based on discriminant-PLS.

At occasions of direct transport over the Baltic sea from one sampling station to another (a distance of approximately 500 kilometres) a relative increase of S and Br and a decrease in Ca, Ti, Mn, Fe and Zn was observed. This is a direct observation of sulphur gas-to-particle conversion taking place during long-range transportation. The increase in particulate Br is believed to emanate from the sea.

The two sampling stations located east and south of Stockholm, the capital of Sweden with approximately 1 million inhabitants, enabled us to study the influence of this city on its surroundings. The results showed that the plume of Stockholm was only clearly discernable when superimposed on clean air masses originating from the Northern Atlantic.

This work was supported by the Swedish National Environmental Protection Board and Stockholms Miljö-och Halsoskyddsforvaltning.

THE INFLUENCE OF AMMONIA FROM AGRICULTURE ON TRENDS IN  
BACKGROUND AEROSOL CONCENTRATIONS

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Due to the increase in agricultural production and livestock numbers, ammonia emissions in Europe are estimated to have increased by about 50% since 1950. This has happened concurrently with changes in anthropogenic emissions of  $\text{SO}_x$  and  $\text{NO}_x$ .

The effect of greater amounts of ammonia is to increase the in-cloud oxidation of sulphur and ammonium aerosol production; however it also increases the capacity of precipitation to deposit sulphur. This paper will raise questions about the net effect of higher ammonia emissions on export of  $\text{SO}_x$  to the free tropospheric circulation by venting aloft from storm systems, and the aerosol concentrations in boundary layer air leaving continental Europe. It will be illustrated using computer modelling studies of selected scenarios.

## ATMOSPHERIC ELECTRICITY IN GALWAY

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More than once it has been tried to discern a trend in the electrical conductivity of the air over the oceans, indicating an increasing background pollution of the atmosphere on a global scale (Cobb and Howard, 1970). But up till now the measurements are too few and the scatter too great for such a trend to be demonstrated beyond doubts. This is due to the following factors:

- A- The systematic decrease of the conductivity is a slow process if it exists at all.
- B- The values for the conductivity were obtained by various techniques.
- C- The results come from various floating locations on the ocean taken under varying weather conditions and in various seasons.

Systematic errors as introduced by B and C can be avoided by measuring the conductivity on a suitable coastal station. A near perfect location for monitoring the background conductivity of North Atlantic air would be the research station at Mace Head, Galway. Optical and chemical properties of the air are already studied there.

Apart from studying this particular trend there is another good reason to establish a station for atmospheric electricity in Galway. The Royal Meteorological Service (KMI) in Belgium runs a small national network of atmospheric electricity stations. The addition of a station at Mace Head would result in a European network at the expense of only a relatively small investment.

Our institute in Utrecht has gained three years of experience in the construction and the continuous operation of atmospheric electricity sensors under severe marine conditions (V.d. Hage, 1988). The present author would gladly put this experience at the disposition of EURASAP by installing an atmospheric electricity station at Mace Head when funds are available, and another one in the Netherlands.

SCAVENGING OF AEROSOL PARTICLES BY DRIZZLE AND PRECIPITATION  
SIZED DROPS

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The efficiency  $E(a,r)$  with which aerosol particles of radius  $r$  ( $> 0.5 \mu\text{m}$ ) are scavenged by a precipitation drop of radius  $a$  (under the simultaneous influence of gravity, drag, electrical forces, thermophoresis and diffusiophoresis) has been determined by means of the trajectory model first used by Grover et al (1977). For aerosol particles with radii  $r < 0.5 \mu\text{m}$  inertial impaction can be ignored and the collision efficiency is determined from the flux model of Wang et al (1978). Reasonable agreement of calculated collision efficiencies using the two models is obtained over the aerosol radius overlap region from 0.1 to 1.0  $\mu\text{m}$ . Collision efficiencies for aerosol-drop interactions were calculated at 900 mb atmospheric pressure and 10 °C ambient temperature for relative humidity values of 50, 75, 95 and 100%. The particle radius ranged from 0.5 to 5  $\mu\text{m}$  whilst the drops radius varied from 50 to 500  $\mu\text{m}$ .

The effect of scavenging of particles by three Marshall- Palmer (M-P) type distributions with precipitation rates of 5, 10.2 and 25 mm/hr<sup>-1</sup>, and duration of 5, 10 and 20 minutes on lognormal aerosol size distributions within radii limits 0.005 to 10  $\mu\text{m}$  is examined for a range of relative humidity and electrical conditions. In addition, the percentage of aerosol particles which are scavenged by precipitation drops possessing M-P distributions parameters as above is calculated.

A THEORETICAL STUDY OF THE WET REMOVAL OF TWO DIFFERENT TYPES  
OF AEROSOL PARTICLES BY MEANS OF A TWO DIMENSIONAL  
DYNAMIC MODEL FOR A CONVECTIVE WARM CLOUD

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A theoretical model has been formulated to study the scavenging of aerosol particles by a warm convective cloud. The model uses detailed microphysics and follows the evolution of the droplet spectra, the aerosol particle spectra in the air and the size distribution of the aerosol particle mass scavenged in the cloud drops at every grid point. The processes considered are activation of aerosol particles (nucleation scavenging), condensation and evaporation of particles and drops, impaction scavenging of aerosol particles through drops, collision and coalescence of drops and breakup of large drops. This microphysical scavenging model is embedded in the two dimensional convective cloud model of Clark et al. Thus, the complete model allows to follow the aerosol mass scavenged during the cloud life time and its deposition onto the ground.

This sophisticated scavenging model has been evaluated for the conditions of day 261 of the GATE experiment.

The initial aerosol particle spectrum was assumed to be of maritime type consisting of a superposition of three log-normal distributions. The nucleation and accumulation mode were assumed to consist of  $(\text{NH}_4)_2\text{SO}_4$  particles and the coarse mode was set to hold only NaCl particles. The particle spectra were assumed to decrease exponentially with height.

On this heterogeneous aerosol particle spectrum cloud drops form through nucleation. When the cloud drops grow they continue to scavenge the two types of particles through impaction. Diffusional growth, collision and coalescence and breakup mix the aerosol particle masses and redistribute them inside the drop spectra so that the main pollution mass is associated with the main water mass. The model allows to trace the location of the two different types of particles inside the drop spectrum in the cloud and in the rain on the ground. The results show reasonable agreement with field observations.

## A FIELD STUDY ON IN-CLOUD REMOVAL OF AEROSOL PARTICLES

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An extensive field campaign was undertaken aimed at observing interactions between aerosol particles and hydrometeors during frontal passages with precipitation in winter time. Observations were made at three ground-based measuring stations along the north-western slope of Mount Rigi, Central Switzerland at 430 - 1620 m a.s.l. The stations were operated simultaneously and were capable of measuring major parameters in and below the clouds. Detailed radar observations of the incoming precipitation completed the observations. Fifteen precipitation systems were observed, the majority of them induced by weak cold fronts. A number of investigations have already been carried out with parts of this data set and have been published recently (Schumann and Waldvogel (1987), Schumann et al. (1988), Zinder et al (1988), Schumann (1989)). This contribution expands the investigations to the study of in-cloud removal processes.

### A) In-cloud scavenging:

Never reported before, size-dependent in-cloud scavenging efficiencies were determined. With the help of such efficiencies in-cloud scavenging could be explained as a combination of pure nucleation scavenging (for 0.2-2  $\mu\text{m}$  particles) and impaction scavenging (by diffusive processes for particles smaller than 0.2  $\mu\text{m}$  and by impaction processes for particles large than 2  $\mu\text{m}$ ). The case-to-case variability of determined efficiencies was very large, being the result of the influence of simultaneous precipitation, the solubility of the aerosols, the number concentration of fine particles ( $d < 0.25 \mu\text{m}$ ), and the maximum supersaturation reached during cloud formation. The experimentally determined efficiencies agree, at least qualitatively, with current nucleation theories e.g. by Hanel (1987). Field experiments reported by other authors reconfirm the mean total efficiency of 70% (by aerosol volume) found in this study.

### B) Precipitation chemistry:

The inorganic ion concentrations found in aerosols and precipitation show that the precipitation first falling contains overproportional amounts of coarse, Ca enriched aerosol particles, indicating that the transformation of cloud water to precipitating water favours droplets originating from particles with pronounced cloud condensation nuclei characteristics. The submicron particles consisting primarily of  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  are quickly incorporated into cloud water, but much less efficiently transformed into precipitating water than super- micron particles. The ion concentrations in below-cloud precipitation are from both in-cloud and below-cloud levels with widely varying percentages.

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A THEORETICAL INVESTIGATION OF THE COLLECTION OF AEROSOL  
PARTICLES BY FALLING ICE CRYSTALS

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A numerical scheme which determines the collection efficiencies, kernels and washout rates of aerosol particles (AP) by falling columnar and platelike ice crystals is presented. A theoretical model for the removal of micron-size APs by falling columnar ice crystals which incorporates gravitational, inertial, thermophoretic, diffusiophoretic and electrostatic forcing has been formulated. This trajectory model which includes computed velocity, temperature and water vapor density fields, was coupled to a flux model which determines the removal of submicron particles due to Brownian diffusion, thermo and diffusio-phoresis as well as electrostatic forcing. This combined model indicates collection efficiencies for APs of radii 0.001 to 10.0 microns for the columnar ice crystal size distribution. An earlier study provides AP collection efficiencies by ice crystal plates for the ice crystal plate size distribution.

The columnar ice crystal-aerosol particle collection model indicates that efficiency increases with increasing pressure, temperature or electrostatic charge and for decreasing relative humidity. An observed zero collection zone near APs of about 2 microns in radius is seen as a near zero balance in the radial forces.

Integrating the product of the computed collection kernels with their appropriate ice crystal distribution will indicate the AP size dependent washout rates. It is seen that ice crystal plates collect APs at significantly greater rates than columnar ice crystals for AP greater than 0.05 microns. APs less than 0.05 microns are collected at the same rate by plates and columns.

The results of this collection rate scheme have been set up as a large data set. This data can be applied to mesoscale air pollution models either directly or in the form of a statistically fit function dependent on ice crystal and aerosol particle characteristic geometries as well as atmospheric pressure, temperature, and relative humidity. This same parameterization technique has also been applied to the collection of APs by raindrops and drizzle. This is the most accurate quantitative approach to computing washout of APs by falling hydrometeors.

THE SCAVENGING OF HIGH ALTITUDE AEROSOL BY SMALL ICE CRYSTALS

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There have been several global models developed for the theoretical investigation of the removal of high altitude aerosol from the atmosphere following concern about the injection of particulate material by nuclear explosions and volcanic events. These models lack a knowledge of the scavenging efficiencies of the small ice crystals associated with cirrus clouds and storm ice anvils. These are the only hydrometers that could act to remove the injected particles.

In the past there have been a number of practical studies into the scavenging efficiencies of large ice crystals and snowflakes. A comparison of the extrapolated results of these findings and the theoretical models of Martin et al (1980) for the small crystal situation has been made. It was found that in general the extrapolated results gave efficiencies that were significantly higher than the predicted value. This difference was found to be enhanced as the crystal diameter decreased.

Experiments using small ice plates grown at  $-18.5^{\circ}\text{C}$  in a cloud chamber which are then permitted to fall through a dense aerosol cloud should provide the first direct measurements of the scavenging efficiencies of these small crystals under cloud conditions.

MODELLING OF WET SCAVENGING OF HEAVY METALS IN THE  
MARINE ENVIRONMENT

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A 3D mesoscale model with parameterized microphysics of clouds and precipitation (Levkov et al., 1988, in print) has been extended to include wet deposition scavenging of heavy metals from the atmosphere. The meteorological input data are derived from the North Sea KonTur experiment (Hoeber, 1982). As a first metal lead has been chosen. Both, calculated (long range transport model), and measured atmospheric monthly mean concentration of lead at the Germany coast of North Sea and Baltic Sea (Petersen et al., 1988) are used as in input pollution level during the simulations with this smaller scale model. The model output data indicate a lead concentration in rain water after 4 hours of simulation between 5 and 10  $\mu\text{g}$  per liter, and a wet deposition rate between 1 and 3  $\mu\text{g}$  per  $\text{m}^2$  per hour. These results show good agreement with measured weekly averages of lead deposition fluxes during precipitation events at Pellworm (Stossel, 1987) selected for validation of the model. Additionally, attention is focussed to demonstrate the nonlinearity between the prognostic variables, the rainfall rate and the atmospheric wet deposition mainly resulting from the lead loading as a function of aerosol particle size. The simulation results obtained with the coupled dynamical, microphysical and cumulus parameterization modules suggest this approach to be an effective tool for studying the heavy metals interaction with the water in the atmospheric environment.

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FIELD MEASUREMENTS OF THE WET DEPOSITION OF PARTICULATE MATERIAL

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Most wet deposition studies have investigated the collection of adventitious trace species. However, these are often better suited to assessing regional pollution budgets rather than contributing to a better understanding of the mechanisms important in leading to wet deposition. Alternative measurements have included plume scavenging studies which have suffered the disadvantage that a size distribution of the scavenged species is only partially known at best. Laboratory studies have been generally criticised because environmental conditions might not be adequately reproduced.

Several releases of particles of known size distribution have been performed during natural precipitation events and washout rates have been assessed. A generalisation of the results has enabled the relative importances of wet deposition via washout and dry deposition rates to be assessed for particulate material. A comparison has been included with theoretical predictions also.

SEQUENTIAL SAMPLING OF MAJOR IONS, DISSOLVED AND TOTAL TRACE METALS IN WET DEPOSITION

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In order to study the processes controlling the removal of aerosols from the atmosphere, a programme of collecting rainwater in remote regions has been instigated. This contribution describes the results of one such experiment at Adrigole, Ireland.

Over a period of 3 days, filtered and unfiltered precipitation were sequentially collected on a wet-only basis in April 1988. During the passage of a frontal system, both stratiform and cumuliform cloud systems were observed and precipitation from both cloud types was collected and analysed for major ions, dissolved and particulate trace metals.

All major ions ( $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{H}^+$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ) showed a decrease in concentration with time, with the exception of ammonium. Elements of marine origin ( $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ) were strongly correlated with each other ( $r: 0.858-0.995$ ) and with the volume of rainfall collected in each fraction. This suggests that these aerosols are susceptible to a dilution effect and may be scavenged as fully wetted aerosols, due to their hydroscopic nature. The rate of wet deposition of ammonium and total sulphate increased with a corresponding change in cloud base level and cloud type. In contrast,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  were independent of rainfall rate and therefore, deposition rate of these elements did not increase with time.

Similarly, dissolved and total trace metal concentrations (Cd, Pb, Mn, Fe, Cu, Zn) decreased over time but the fall off rate was much less rapid than for major ions. In contrast to most major ions, there was no volume dependency and therefore no dilution effect was observed for these aerosols. The rate of deposition of dissolved Cu, Pb, Cd and for all total trace metals increased with a change in cloud system.

In general, hydrogen ion concentration varied in phase with the trace metals measured. Correlations with dissolved metals were higher than with total metals.

DAILY MEASUREMENTS OF BACKGROUND AIR POLLUTANTS AT A SITE  
IN THE WEST OF NORTHERN IRELAND

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Daily measurements of precipitation composition, SO<sub>2</sub> and aerosol sulphate concentration have been made at Lough Navar in the West of Northern Ireland from 1986 to 1988. Rainwater was collected using an automatic wet only collector and aerosol concentrations measured using a dichotomous sampler.

Using a trajectory analyses, measurements were assigned to one of four categories: Atlantic, Ireland, UK/Europe and undefined. The annual mean concentrations associated with Atlantic trajectories were used as estimates of the background concentrations. The variability of the background was determined from the frequency distributions of the daily concentrations and wet deposition associated with Atlantic trajectories.

The simultaneous sampling of aerosol and rainwater has allowed scavenging in marine air masses to be investigated and results will be presented.

SEASONAL VARIATIONS IN SULFATE, NITRATE, AND CHLORIDE IN  
THE GREENLAND ICE SHEET: RELATION TO ATMOSPHERIC CONCENTRATIONS

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Samples from two snowpits near Dye 3 in south Greenland have been used to study seasonal variations in contaminant transport from the atmosphere to the ice Sheet. The samples have been dated by comparing  $\delta O^{18}$  values with meteorological data from Dye 3. Airborne concentrations over the Ice Sheet have been estimated for the dates corresponding to each snowpit sample by statistically analyzing data from several air monitoring stations throughout the Arctic, and computing average values from the appropriate stations. Seasonal variations in concentrations in air, concentrations in snow, and mass-basis scavenging ratios (concentration in air divided by concentration in snow) have been identified. Results indicate that concentrations of  $SO_4^{2-}$  in the air show a strong peak in late February, while those in the snow show a weaker peak in early March with more uniform concentrations throughout the year. The smaller seasonal variation in the snow is attributed to the effect of riming, which results in more efficient scavenging during warmer weather. In agreement with previous estimates, dry deposition appears to account for 10 - 30% of the total  $SO_4^{2-}$  in the snow. Concentrations of  $NO_3^-$  in the air peak in late February while those in the snow peak in mid-August, reflecting the influence of riming and possible high altitude sources of  $NO_3^-$ . Concentrations of  $Cl^-$  in the air and snow each show a winter peak and a summer peak. The former reflects long-range transport of marine aerosol at a time when seas in the Arctic are frozen, while the latter results from local seaspray. Scavenging ratios for  $SO_4^{2-}$  and  $NO_3^-$  peak in early August while those for  $Cl^-$  peak in early June; these variations in part reflect more efficient scavenging during warm weather.

## AUTHOR INDEX

	Page
Anacleto, T.D.	31
ApSimon, H.M.	86
Ayatollahi, Sh.	75
Baltensperger, U.	47
Bell, D.A.	93
Below, M.	24
Bergametti, G.	72, 73, 74
Bigg, E.K.	5
Borbély-Kiss, I.	30
Borowska, M.	58
Bowdle, D.A.	2, 42
Bowdle, D.A.	44, 62
Brand, P.	24
Branson, J.R.	95
Bremond, M.P.	34
Buat-Ménard, P.	34, 84
Bunz, H.	55
Burgermeister, S.	28
Butterweck, G.	74
Cachiér, M.P.	34
Carlier, P.	72, 73
Castillo, R.	32
Chýlek, P.	61
Clarke, A.D.	26, 44, 45
Consterdine, I.E.	22
d'Almeida, G.A.	39
Davidson, C.I.	90
Devara, P.C.S.	67
Dreiling, V.	23
Dybizbanski, J.	58
Eppel, D.P.	94
Eyles, N.A.	56
Fernandez, G.	81
Fitzgerald, J.W.	7
Fisher, G.W.	20, 79
Flossmann, A.I.	89
Fytianos, K.	51
Gaggeler, H.W.	47
Galloway, J.N.	82
Gardneus, L.	80
Garvey, D.M.	81
Gebhart, J.	24
Geiss, F.	51
Georgi, B.	24, 49, 50
Georgii, H.W.	28
Gras, J.L.	4
Grassl, H.	83, 94
Hameri, K.	68
Hamilton, R.S.	37
Hansen, A.D.A.	36
Hansen, C.	69
Hansson, H.C.	85
Harrison, R.M.	29
Harve, M.J.	20, 79

	Page
Heintzenberg, J.	35
Hess, M.	39
Heyder, J.	24
Hitzenberger, R.	80
Holt, E.H.	27, 81
Horvath, H.	41
Huebert, B.J.	57
Hummelshoj, P.	69
Isaac, P.	20
Jaenicke, R.	77
Jennings, S.G.	21, 25
Jennings, S.G.	27, 61, 88
Jickells, T.D.	96
Joffre, S.M.	71
Jost, D.T.	47
Kahl, J.D.	3
Kaye, A.D.	5, 30
Keeler, G.	32
Kharchilava, D.F.	52
Koepke, P.	39
Koltai, E.	30
Konttinen, S.	53
Kotzias, D.	51
Koyra, M.	55
Knobben, R.A.	79
Kruse-Plass, M.	86
Kulmala, M.	68
Larsen, H.R.	79
Laube, M.	70
Leaitch, W.R.	78
Leal, R.M.	31
Lechner, I.S.	20, 79
Lee, G.	57
Levkov, L.	94
Lim, B.	96
Lindfors, B.	71
Lomaya, O.V.	52
Losno, R.	72, 73, 84
Ludlow, I.K.	56
Mansfield, T.A.	37
Maser, R.	23
Mészáros, E.	30
McCormick, M.P.	40
McGann, B.T.	88
McGovern, F.M.	6, 21
Megaw, J.W.	5, 38
Menzies, R.T.	42
Miller, N.L.	92
Molnár, Á.	30
Mouvier, G.	72
Müller, H.-N.	50
Muerer, M.	50
Nicholson, K.W.	95
O'Connor, T.C.	6, 21

	Page
O'Dowd, C.D.	21, 25, 27
Ogren, J.A.	80
Pacyna, J.	83
Park, P.M.	22
Pastuszka, J.S.	58
Patterson, E.M.	44
Petersen, G.	83
Pinnick, R.G.	27, 81
Pio, C.A.	31
Porstendorfer, J.	74
Post, M.J.	42, 44
Post, M.J.	62, 63
Preining, O.	1
Pruppacher, H.R.	89
Pueschel, R.F.	40
Raj, P.E.	67
Raunemaa, T.	53, 68
Reineking, A.	74
Reist, P.C.	64
Remoudaki, E.	84
Rogers, C.F.	59
Rothermel, J.	42
Russell, P.B.	40
Santos, I.M.	31
Saunders, C.P.R.	93
Scheidgen, P.	70
Schnell, R.C.	3, 36
Schock, W.	55
Schumann, T.	90
Schutz, L.	23
Serpolay, R.	60
Smith, M.H.	32
Sommer, H.T.	33
Spengler, J.	32
Stedman, J.	97
Stott, P.	86
Strom, J.	80
Svantesson, B.	85
Swietlicki, E.	30
Szabo, G.	65
Sztyler, A.	75
Taheri, M.	68
Tapper, U.	66
van Daalen, J.	87
van der Hage, J.C.H.	42
Vaughan, J.M.	57
Warren, W.	83
Weber, H.	82
Whelpdale, D.M.	86
Whitecombe, G.	64
Wilson, W.	19
Wu, J.	68
Ylatalo, S.	61
Zhan, J.	54
Zwozdziak, J.W.	